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Universal ratio of TTLS-phonon coupling constants in low-temperature amorphous solids

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Abstract

Tunneling-two-level-system model (TTLS model) has successfully explained several universal properties of amorphous solids at low temperatures. The experimentalists found that the ratios of TTLS-phonon coupling constants γ_l/γ_t lie between 1.44 and 1.84 among 16 different amorphous solids, which turns out to be a universal quantity (Berret and Meissner 1988 *Z. Phys. B* **70** 65). However, this universal property remains unexplained. Based on the model introduced by Vural and Leggett (2011 *J. Non-Cryst. Solids* **357** 3528) and real space renormalization, we demonstrate that γ_l/γ_t equals to c_l/c_t , where c_l and c_t are longitudinal and transverse sound velocities, respectively. In this paper, we reveal that this universal quantity γ_l/γ_t essentially comes from the mutual interactions between elementary blocks of amorphous solids, and is insensitive to the microscopic material properties. In the appendix, we also make corrections to the many-body interaction of low-temperature amorphous solids derived by Joffrin and Levelut (1975 *J. Physique* **36** 811).

Keywords: glass, universality, low temperature

(Some figures may appear in colour only in the online journal)

1. Introduction

It has been more than 40 years since Zeller and Pohl discovered [4] that below $T = 1$ K, the thermal and acoustic properties of amorphous solids (glasses) behave strikingly universal and different from that of the crystalline counterparts. Later, Anderson [5] *et al* and Phillips [6] independently developed a microscopic phenomenological model which was later known as tunneling-two-level-system (TTLS) model. It successfully explained several universal properties, such as linear-temperature dependence of heat capacity, phonon saturation [7], echoes [8, 9], and quadratic-temperature dependence of heat conductivity [10, 11], etc.

In TTLS model a group of tunneling-two-level-systems (TTLSs) are randomly embedded in the amorphous solid. The effective Hamiltonian of amorphous solid is the summation

of long wavelength phonon Hamiltonian, the Hamiltonian of TTLSs, and the coupling between phonon strain fields and TTLSs. The coupling constant between longitudinal (transverse) phonon strain and TTLSs is denoted as γ_l (γ_t). γ_l and γ_t are freely adjustable parameters and are independent of each other in TTLS model.

However, later experiments suggested that γ_l and γ_t are not arbitrary, and the ratio γ_l/γ_t turns out to be quite universal. In 1988, Meissner and Berret [1] summarized the measurements of γ_l and γ_t from 16 different amorphous materials, including chemically pure materials (e.g. a-SiO₂), chemically mixed materials (e.g. BK7) and organic materials (e.g. PMMA). They found that γ_l/γ_t ranges from 1.44 to 1.84 among these materials. Moreover, most of the ratios are around 1.5–1.6. Such significant universality cannot be explained within TTLS model [12] since the model itself depends on these coupling

constants. Intrigued by the lack of theoretical explanation, we propose that there should be a more general model for the low-temperature universal properties of amorphous solids, including the universal ratio γ_l/γ_t .

TTLSs are coupled to phonon strain fields and can resonantly absorb phonon energy when the energy splitting of TTLS matches phonon frequency. Based on Fermi's golden rule, the energy absorption rate of longitudinal (transverse) phonon is $dE_l/dt \propto \gamma_l^2 (dE_t/dt \propto \gamma_t^2)$, which means $(dE_l/dt)/(dE_t/dt) = \gamma_l^2/\gamma_t^2$. The universal ratio γ_l/γ_t therefore is equivalent to the universal ratio $(dE_l/dt)/(dE_t/dt)$ in TTLS model.

In this paper, we conduct theoretical analysis on longitudinal and transverse phonon resonance energy absorption of amorphous solids. We aim to prove that the ratio $(dE_l/dt)/(dE_t/dt)$ is universal in amorphous solids. By referring to the model of Vural and Leggett [2], we consider the general Hamiltonian \hat{H}^{tot} of an elementary block of amorphous solid. The Hamiltonian can be expressed as the summation of long wavelength phonon Hamiltonian \hat{H}^{el} (i.e. purely elastic Hamiltonian) and the rest part $\hat{H}^{\text{tot}} - \hat{H}^{\text{el}}$. Here, we denote $\hat{H}^{\text{non}} \stackrel{\text{def}}{=} \hat{H}^{\text{tot}} - \hat{H}^{\text{el}}$, which is termed as 'non-harmonic part of glass Hamiltonian'. We denote e_{ij} as the strain field of long wavelength phonon, where the lower indices $i, j = 1, 2, 3$ label the cartesian coordinates. We then expand \hat{H}^{non} in orders of the strain field, e_{ij} . The coefficient of the first order expansion, $\hat{T}_{ij}^{\text{non}}(\vec{x}) \stackrel{\text{def}}{=} \partial \hat{H}^{\text{non}} / \partial e_{ij}(\vec{x})$, is named as 'non-harmonic stress tensor'. Similar to the coupling between TTLSs and phonon strain fields, the non-harmonic stress tensors are coupled to phonon strain as well.

In what follows, we allow virtual phonons between different elementary blocks to exchange with each other to generate an additional many-body interaction. This is a long-range interaction with $|\vec{x} - \vec{x}'|^{-3}$ behavior between non-harmonic stress tensors at different positions and therefore is the mutual interaction between elementary blocks. We then combine elementary blocks to form a super block. The Hamiltonian of the super block is the summation of elementary block Hamiltonian and the mutual interaction between elementary blocks. Based on this Hamiltonian, we carry out the recursion relation of resonance phonon absorption between super block Hamiltonian and elementary block Hamiltonian. This is one step of real space renormalization of phonon energy absorption. It exhibits how phonon energy absorption changes as the length scale increases. Initiating from the microscopic length scale, we repeat the renormalization process until the experimental length scale is reached. The ratio of energy absorption $(dE_l/dt)/(dE_t/dt)$, eventually flows to $(c_l/c_t)^2$ at experimental length scale where c_l and c_t are longitudinal and transverse sound velocities, respectively. As we have noted that γ_l^2/γ_t^2 equals to $(dE_l/dt)/(dE_t/dt)$, the ratio of coupling constants γ_l/γ_t equals to c_l/c_t at experimental length scale. The ratio of sound velocities c_l/c_t is an experimentally measurable quantity with no adjustable parameters. We believe the universal ratio γ_l/γ_t essentially comes from the mutual interactions between elementary blocks, so it is insensitive to the microscopic structures and chemical compositions of the

amorphous materials, which we will discuss in more detail in section 4.

The paper is organized as follows. In section 2 we briefly review TTLS model and explain how the problem of universal ratio γ_l/γ_t arises. We then introduce the generalized model of amorphous solids [2], along with the concepts of non-harmonic part of glass Hamiltonian, non-harmonic stress tensor and non-harmonic susceptibility. By allowing the exchange of virtual phonons, we derive the many-body interaction to further obtain the full Hamiltonian of super block amorphous solid. In section 3 we perform a detailed calculation on the resonance phonon energy absorption based on the above general Hamiltonian. We set up the real space renormalization equation of energy absorption between small and large length scales of the Hamiltonians. In section 4 we repeat the renormalization process from the microscopic length scale to experimental length scale and achieve the universal ratio between longitudinal and transverse phonon energy absorptions. In section 5 we compare our theory with experimental data of γ_l/γ_t and discuss the statistical significance between them for 16 amorphous materials. In appendix A we provide a detailed correction to the non-harmonic stress-stress many-body interaction, which was originally derived by Joffrin and Levelut [3].

2. The model

2.1. Short review of TTLS model

Let us briefly review the ratio γ_l/γ_t and TTLS model [5, 6] in this subsection.

We first define $e_{ij}(\vec{x})$, the intrinsic strain field of long wavelength phonon. Later we will introduce externally applied strain field $e_{ij}(\vec{x}, t)$ when considering resonance phonon energy absorption process. Given that $\vec{u}(\vec{x})$ is the relative displacement of the matter at position \vec{x} , the intrinsic strain field $e_{ij}(\vec{x})$ can be written as

$$e_{ij}(\vec{x}) = \frac{1}{2} \left(\frac{\partial u_i(\vec{x})}{\partial x_j} + \frac{\partial u_j(\vec{x})}{\partial x_i} \right), \quad (2.1)$$

where $i, j = 1, 2, 3$, and x_i, x_j stand for cartesian coordinates. By decomposing displacement field $\vec{u}(\vec{x})$ into longitudinal and transverse parts, we divide the strain field e_{ij} as the longitudinal part $e_{ij,l}$ and the transverse part $e_{ij,t}$.

According to TTLS model, a group of TTLSs are randomly distributed in the amorphous material. The effective Hamiltonian \hat{H}^{tot} is expressed as the summation of three terms,

$$\hat{H}^{\text{tot}} = \hat{H}^{\text{el}} + \hat{H}^{\text{TTLS}} + \hat{H}^{\text{coup}}. \quad (2.2)$$

The first term \hat{H}^{el} is the purely elastic Hamiltonian (i.e. long wavelength phonon Hamiltonian)

$$\hat{H}^{\text{el}} = \sum_k \sum_{\alpha=l,t} \hbar \omega_{k\alpha} (\hat{a}_{k\alpha}^\dagger \hat{a}_{k\alpha} + 1/2) \quad (2.3)$$

where k is the phonon wave number, and $\alpha = l, t$ denote longitudinal phonon and transverse phonon, respectively. The second term \hat{H}^{TTLS} is the Hamiltonian of TTLSs,

$$\hat{H}^{\text{TTLS}} = \sum_{N=1}^{N_{\text{tot}}} \frac{1}{2} \begin{pmatrix} E_N & 0 \\ 0 & -E_N \end{pmatrix}, \quad (2.4)$$

where the TTLS Hamiltonian is given in the basis of its energy eigenstates with the energy splitting $E_N = \sqrt{\Delta_N^2 + \Delta_{0N}^2}$, Δ_N is the asymmetry of the double-well potential, Δ_{0N} is the tunneling splitting, and N_{tot} is the number of TTLSs in the amorphous solid. The third term in equation (2.2) is the coupling between TTLSs and phonon strain fields,

$$\hat{H}^{\text{coup}} = \sum_{\alpha=l,t} \sum_{N=1}^{N_{\text{tot}}} \frac{\gamma_{\alpha} e_{\alpha}(\vec{x}_N)}{2} \begin{pmatrix} D_N & M_N \\ M_N & -D_N \end{pmatrix}. \quad (2.5)$$

In the original literatures of the TTLS model, the tensorial nature of the phonon strain field has been ignored, and $\gamma_l e_l(\vec{x}_N)$ ($\gamma_t e_t(\vec{x}_N)$) is written as the coupling between TTLSs and orientation-averaged longitudinal (transverse) phonon strain fields [6, 13, 14]. Thus, $e_l(\vec{x}_N)$ ($e_t(\vec{x}_N)$) is the orientation-averaged longitudinal (transverse) component of phonon strain field which couples to the N th TTLS, and γ_l (γ_t) is the coupling constant for the longitudinal (transverse) phonon fields. $D_N = \Delta_N/E_N$ and $M_N = \Delta_{0N}/E_N$ are the normalized matrix elements of the TTLS-phonon couplings [13, 15]. D_N and M_N are in general different for longitudinal and transverse phonon strain fields, but usually they are assumed to be equal based on the assumption that the strain fields mainly couple to the asymmetry Δ_N [13].

Having established TTLS model, we now define TTLS resonance susceptibilities as follows,

$$\chi_{\alpha}(\vec{x}_N, \vec{x}'_N; \omega)|_{\text{TTLS}} \stackrel{\text{def}}{=} -\frac{\delta}{\delta e_{\alpha}(\vec{x}'_N)} \left\langle \frac{\delta(\hat{H}^{\text{tot}} - \hat{H}^{\text{el}})}{\delta e_{\alpha}(\vec{x}_N)} \right\rangle, \\ \chi_{\alpha}(\omega)|_{\text{TTLS}} \stackrel{\text{def}}{=} \frac{1}{N_{\text{tot}}} \sum_{N,N'=1}^{N_{\text{tot}}} \chi_{\alpha}(\vec{x}_N, \vec{x}'_N; \omega)|_{\text{TTLS}}, \quad (2.6)$$

where $\langle \dots \rangle$ represents thermal average and quantum average [2], and $\chi_{\alpha}(\vec{x}_N, \vec{x}'_N; \omega)|_{\text{TTLS}}$ and $\chi_{\alpha}(\omega)|_{\text{TTLS}}$ are TTLS resonance susceptibility and space-averaged susceptibility, respectively. This definition is in accordance with [6, 13, 14, 16], except for the negative sign of the definition of TTLS resonance susceptibility we made in equation (2.6).

The imaginary part of the space-averaged TTLS susceptibility is obtained from equation (2.6) by employing linear response theory,

$$\text{Im } \chi_{\alpha}(\omega)|_{\text{TTLS}} \\ = \frac{1}{N_{\text{tot}}} \sum_{N=1}^{N_{\text{tot}}} \frac{\pi M_N^2 \gamma_{\alpha}^2}{4} \tanh\left(\frac{\beta \hbar \omega}{2}\right) \delta(E_N - \hbar \omega), \quad (2.7)$$

where T is the temperature, $\beta = 1/k_B T$, $Z_N = 1 + e^{-\beta E_N}$ is the partition function, and the factor $\tanh(\beta \hbar \omega/2) = (1 - e^{-\beta E_N})/Z_N$ is the population difference between two levels. A detailed calculation of equation (2.7) is obtained by Hunklinger and Arnold [14] which is analogous to the susceptibility of spin system in response to magnetic fields. Note that this imaginary part of susceptibility is positive due to the negative sign we defined in equation (2.6). From

equation (2.7) it is worth noticing that the frequency dependence of $\text{Im } \chi_l(\omega)|_{\text{TTLS}}$ is the same as that of $\text{Im } \chi_t(\omega)|_{\text{TTLS}}$, i.e. both of them are proportional to $\tanh(\beta \hbar \omega/2)$:

$$\text{Im } \chi_{\alpha=l,t}(\omega)|_{\text{TTLS}} \propto \tanh(\beta \hbar \omega/2). \quad (2.8)$$

Equation (2.8) is essential to perform the renormalization argument in section 4.

The real part of space-averaged TTLS susceptibility was first derived by Hunklinger and Arnold [14],

$$\text{Re } \chi_{\alpha}(\omega)|_{\text{TTLS}} \\ = -\frac{n_0 M^2}{4} \left[\text{Re } \Psi \left(\frac{1}{2} + \frac{\beta \hbar \omega}{2\pi i} \right) + \ln \left(\frac{1}{\beta \hbar \omega} \right) \right], \quad (2.9)$$

where the overall negative sign arises from the negative sign in equation (2.6). M is the off-diagonal element of the TTLS-phonon coupling, and is assumed to be constant for two-level-systems in the literatures [6, 13, 14]. The overall factor of 1/4 originates from the definition of M in equation (2.5), where $2M$ is adopted by Hunklinger and Arnold [14]. n_0 is the constant density of states of two-level-systems, and Ψ is the digamma function. Equation (2.9) was used by Phillips [6] and Piché *et al* [16] to explain the measurements of sound velocity shift in amorphous solids. Ultrasonic measurements [1, 14, 16] with the parameters $\omega \sim 10^6$ rad s^{-1} and $T \sim 1$ K are in the region $\beta \hbar \omega \sim 10^{-5} \ll 1$. Thus, the real part of space-averaged TTLS susceptibility is simplified as follows [14],

$$\text{Re } \chi_{\alpha}(\omega)|_{\text{TTLS}} = -\frac{n_0 M^2}{4} \ln \left(\frac{1}{\beta \hbar \omega} \right) < 0. \quad (2.10)$$

Equation (2.10) plays an important role in real space renormalization in section 4. It indicates that within the physical parameters of ultrasonic measurements, the real part of space-averaged susceptibility is negative in TTLS model.

TTLSs can resonantly absorb external phonon wave when the energy splitting E_N matches phonon frequency $\hbar \omega$. Given a monochromatic traveling plane wave $\vec{u}(\vec{x}, t) = \vec{A} e^{i\vec{k}\cdot\vec{x} - i\omega t}$, we shall get the corresponding strain field as $e_{ij}(\vec{x}, t) = \frac{1}{2} (A_i k_j + A_j k_i) e^{i\vec{k}\cdot\vec{x} - i\omega t}$ through equation (2.1), where $\vec{A} = (A_1, A_2, A_3)$ and $\vec{k} = (k_1, k_2, k_3)$ are the phonon vibration vector and the wave vector, respectively. The resonance energy absorption rate is proportional to the coupling constant squared:

$$\frac{dE_{l,t}}{dt} = 2N_{\text{tot}} A^2 k^2 \omega \text{Im } \chi_{\alpha}(\omega)|_{\text{TTLS}} \\ \Rightarrow \frac{dE_l/dt}{dE_t/dt} = \frac{\gamma_l^2}{\gamma_t^2}. \quad (2.11)$$

According to Fermi's golden rule, equation (2.11) can be derived by time-dependent perturbation theory on the coupling between TTLSs and external strain fields. Detailed calculations of equation (2.11) are carried out by both of Jackle [15] and Phillips [6] on the TTLS-phonon scattering rate.

If γ_l and γ_t are freely adjustable parameters and are independent of each other for different amorphous materials, there should be no specific relation between dE_l/dt and dE_t/dt .

In other words, given arbitrary values of γ_l and γ_t , the ratios γ_l/γ_t and $(dE_l/dt)/(dE_t/dt)$ cannot be universal.

However, in 1988 Meissner and Berret [1] summarized the measurements of γ_l and γ_t from 16 amorphous materials, including chemically pure materials (e.g. a-SiO₂), chemically mixed materials (e.g. BK7) and organic materials (e.g. PMMA and PS). Interestingly they found that γ_l/γ_t is not arbitrary. It lies between 1.44 and 1.84 and most of the data are around 1.5–1.6. We note that TTLS model relies on these parameters and it cannot explain this remarkably universal ratio.

From equation (2.11) we can explain the universal γ_l/γ_t by studying the ratio of resonance phonon energy absorption $(dE_l/dt)/(dE_t/dt)$ of the Hamiltonian of amorphous solid. The idea is to introduce the model from Vural and Leggett [2], so that we can present a more general approach to explain the universal ratio $(dE_l/dt)/(dE_t/dt)$.

It is also worth pointing out that many of the γ_l/γ_t data from the original literature were not measured by resonance phonon energy absorption, i.e. the imaginary part of resonance TTLS susceptibility $\text{Im } \chi_{\alpha=l,t}(\omega)|_{\text{TTLS}}$. In fact, Meissner and Berret [1] summarized the measurements of the relative sound velocity shift $\Delta c_{\alpha=l,t}(T)/c_{\alpha=l,t}(T)$ in resonance interacting process for the amorphous materials, which can be expressed as the ratio of *real* part of resonance TTLS susceptibility, i.e. $\gamma_l/\gamma_t = \sqrt{\text{Re } \chi_l(\omega)|_{\text{TTLS}}/\text{Re } \chi_t(\omega)|_{\text{TTLS}}}$. At first glance, resonance energy absorption (i.e. *imaginary* part of TTLS susceptibility $\text{Im } \chi_{\alpha=l,t}(\omega)|_{\text{TTLS}}$) is not reasonable enough for the explanation of γ_l/γ_t . If we refer to Kramers–Kronig relation, the imaginary part of susceptibility can be converted into the real part by integrating over the frequencies. The frequency dependence of the imaginary part of susceptibility may affect the absolute value of the real part, but should not affect the ratio between $\text{Re } \chi_l(\omega)|_{\text{TTLS}}$ and $\text{Re } \chi_t(\omega)|_{\text{TTLS}}$. We will discuss this in detail in section 4. Again we attempt to explain the universal γ_l/γ_t ratio by calculating the ratio of resonance phonon energy absorption, $(dE_l/dt)/(dE_t/dt)$ of amorphous solids.

2.2. Non-harmonic Hamiltonian, stress tensor and susceptibility of amorphous solids

To introduce the model proposed by Vural and Leggett [2], we first generalize TTLS Hamiltonian to multiple-level-system Hamiltonian. Let us consider a block of amorphous material with the side length $L \gg a$, where $a \sim 3 \text{ \AA}$ is the atomic distance. We denote the Hamiltonian of this elementary block as \hat{H}^{tot} . First of all, \hat{H}^{tot} must contain purely elastic Hamiltonian (i.e. long wavelength phonon Hamiltonian \hat{H}^{el} , as shown in equation (2.3)). By subtracting the purely elastic Hamiltonian, we name the leftover part $\hat{H}^{\text{tot}} - \hat{H}^{\text{el}}$ as ‘the non-harmonic part of Hamiltonian’. We further denote it as $\hat{H}^{\text{non}} \stackrel{\text{def}}{=} \hat{H}^{\text{tot}} - \hat{H}^{\text{el}}$.

We then expand the non-harmonic Hamiltonian \hat{H}^{non} with respect to strain field. Up to the first order of intrinsic strain field, the expansion is given as follows,

$$\hat{H}^{\text{tot}} = \hat{H}^{\text{el}} + \hat{H}^{\text{non}}$$

$$\hat{H}^{\text{non}} = \hat{H}_0^{\text{non}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}) \frac{\delta \hat{H}^{\text{non}}}{\delta e_{ij}(\vec{x})} + \mathcal{O}(e_{ij}^2). \quad (2.12)$$

We term the coefficient of the first order expansion as the non-harmonic stress tensor,

$$\frac{\delta \hat{H}^{\text{non}}}{\delta e_{ij}(\vec{x})} \stackrel{\text{def}}{=} \hat{T}_{ij}^{\text{non}}(\vec{x}). \quad (2.13)$$

Now we compare equation (2.12) with TTLS Hamiltonian given in equation (2.2): \hat{H}_0^{non} is the multiple-level generalization of TTLS Hamiltonian \hat{H}^{TTLS} ; $e_{ij}(\vec{x})\hat{T}_{ij}^{\text{non}}$ is the generalization of TTLS-phonon coupling \hat{H}^{coup} .

We denote $\{|m\rangle\}$ and $\{E_m\}$ to be the complete and orthogonal set of eigenstates and corresponding eigenvalues of \hat{H}_0^{non} . Next we define the most important quantity throughout this paper, namely the ‘non-harmonic susceptibility’. Let us impose an external infinitesimal testing strain, $e_{ij}(\vec{x}, t) = e_{ij} e^{i(\vec{k}\cdot\vec{x} - \omega t)}$ to the material. The amorphous material responds with a vibrational stress $\langle \hat{T}_{ij}^{\text{non}} \rangle(\vec{x}, t) = \langle \hat{T}_{ij}^{\text{non}} \rangle e^{i(\vec{k}\cdot\vec{x} - \omega t)}$. Here, the ‘non-harmonic susceptibility [17]’, $\chi_{ijkl}^{\text{non}}(\vec{x}, \vec{x}'; t, t')$ is defined as follows,

$$\chi_{ijkl}^{\text{non}}(\vec{x}, \vec{x}'; t - t') \stackrel{\text{def}}{=} -\frac{\delta \langle \hat{T}_{ij}^{\text{non}} \rangle(\vec{x}, t)}{\delta e_{kl}(\vec{x}', t')}$$

$$\chi_{ijkl}^{\text{non}}(\vec{x}, \vec{x}'; \omega) = \int dt dt' e^{i\omega(t-t')} \chi_{ijkl}^{\text{non}}(\vec{x}, \vec{x}'; t - t'). \quad (2.14)$$

Note that our sign conventions in equations (2.12) and (2.14) makes the eigenvalues of the positive-frequency imaginary part of non-harmonic susceptibility positive. From now on, we always omit the upper indices of \hat{H}^{non} , \hat{H}_0^{non} , χ_{ijkl}^{non} and $\hat{T}_{ij}^{\text{non}}$ for simplicity and denote them as \hat{H} , \hat{H}_0 , χ_{ijkl} and \hat{T}_{ij} .

We further define the space-averaged non-harmonic susceptibility for a block of amorphous material with the volume $V = L^3$:

$$\chi_{ijkl}(\omega) \stackrel{\text{def}}{=} \frac{1}{L^3} \int d^3x d^3x' \chi_{ijkl}(\vec{x}, \vec{x}'; \omega). \quad (2.15)$$

We assume that the amorphous material is isotropic and therefore is invariant under real space SO(3) rotational group. The space-averaged non-harmonic susceptibility $\chi_{ijkl}(\omega)$ yields the isotropic form: $\chi_{ijkl} = (\chi_l - 2\chi_t)\delta_{ij}\delta_{kl} + \chi_t(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$, where χ_l is the compression modulus and χ_t is the shear modulus. From equations (2.14) and (2.15), the imaginary part of $\chi_{ijkl}(\omega)$ is given as follows,

$$\text{Im } \chi_{ijkl}(\omega) = (1 - e^{-\beta\omega}) \text{Im } \tilde{\chi}_{ijkl}(\omega),$$

$$\text{Im } \tilde{\chi}_{ijkl}(\omega) = \sum_m \frac{\pi}{L^3} P_m \int d^3x d^3x' \sum_n \langle m | \hat{T}_{ij}(\vec{x}) | n \rangle \langle n | \hat{T}_{kl}(\vec{x}') | m \rangle \delta(E_n - E_m - \omega), \quad (2.16)$$

where $P_m = e^{-\beta E_m}/\mathcal{Z}$ is the probability function of the m th state, $\mathcal{Z} = \sum_n e^{-\beta E_n}$ is the partition function of \hat{H}_0 , and we set $\hbar = 1$. $\text{Im } \tilde{\chi}_{ijkl}(\omega)$, namely ‘imaginary part of reduced non-harmonic susceptibility’, is defined from equation (2.16). It also yields the isotropy,

$$\text{Im } \tilde{\chi}_{ijkl}(\omega) = [\text{Im } \tilde{\chi}_l(\omega) - 2 \text{Im } \tilde{\chi}_t(\omega)] \delta_{ij} \delta_{kl} + \text{Im } \tilde{\chi}_t(\omega) (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}), \quad (2.17)$$

where $\text{Im } \tilde{\chi}_{\alpha=l,t}(\omega) = \text{Im } \chi_{\alpha=l,t}(\omega)/(1 - e^{-\beta\omega})$. We underline that for $\omega > 0$, $\text{Im } \tilde{\chi}_{\alpha=l,t}(\omega) > 0$. This is the result of the sign convention given in equation (2.14).

Finally, let us discuss the frequency dependence of $\text{Im } \chi_l(\omega)$, $\text{Im } \chi_r(\omega)$ and $\text{Im } \tilde{\chi}_l(\omega)$, $\text{Im } \tilde{\chi}_r(\omega)$ of the multiple-level-system. According to equation (2.8), both of $\text{Im } \chi_l(\omega)|_{\text{TTLs}}$ and $\text{Im } \chi_r(\omega)|_{\text{TTLs}}$ are proportional to $\tanh(\beta\hbar\omega/2)$, i.e. their frequency dependences are the same. However, in our model the frequency dependences of $\text{Im } \chi_l(\omega)$ and $\text{Im } \chi_r(\omega)$ are unknown, because the matrix elements of stress tensors are not specified. In order to proceed the renormalization argument in section 4, we assume that the frequency dependences of $\text{Im } \chi_l(\omega)$ and $\text{Im } \chi_r(\omega)$ (as well as $\text{Im } \tilde{\chi}_l(\omega)$ and $\text{Im } \tilde{\chi}_r(\omega)$) are roughly the same.

2.3. Many-body interaction

In the last subsection, we consider the amorphous solid within a single elementary block with the side length L . The quantities \hat{H}_0 , $\hat{T}_{ij}(\vec{x})$ and $\chi_{ijkl}(\omega)$ we defined are simply generalizations from TTLs to multiple-level-system. So far we cannot acquire anything nontrivial beyond TTLs model. However, if we combine a set of elementary blocks together to form a ‘super block’, the interactions between elementary blocks must be considered and they significantly affect the physical properties of amorphous solids. Given that $\hat{T}_{ij}(\vec{x})$ is coupled to intrinsic strain $e_{ij}(\vec{x})$, if we allow virtual phonons to exchange with each other, the virtual phonon exchange process will generate a many-body interaction between the stress tensors at different positions:

$$\hat{V} = \int d^3x d^3x' \sum_{ijkl} \Lambda_{ijkl}(\vec{x} - \vec{x}') \hat{T}_{ij}(\vec{x}) \hat{T}_{kl}(\vec{x}'), \quad (2.18)$$

where $\hat{T}_{ij}(\vec{x})$ is the stress tensor of elementary block. The coefficient $\Lambda_{ijkl}(\vec{x} - \vec{x}')$ was originally derived by Joffrin and Levelut [3]. The form of their result is in general correct, but we find some of the coefficients are in error. Hence, we obtain a corrected version in equation (2.19), which is derived explicitly in appendix A.

$$\begin{aligned} \Lambda_{ijkl}(\vec{x} - \vec{x}') &= -\frac{\tilde{\Lambda}_{ijkl}(\vec{n})}{8\pi\rho c_l^2 |\vec{x} - \vec{x}'|^3} \\ \tilde{\Lambda}_{ijkl} &= \frac{1}{4} \left\{ (\delta_{jl} - 3n_j n_l) \delta_{ik} + (\delta_{jk} - 3n_j n_k) \delta_{il} \right. \\ &\quad \left. + (\delta_{ik} - 3n_i n_k) \delta_{jl} + (\delta_{il} - 3n_i n_l) \delta_{jk} \right\} \\ &\quad + \frac{1}{2} \left(1 - \frac{c_t^2}{c_l^2} \right) \left\{ -(\delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{jk} \delta_{il}) \right. \\ &\quad \left. + 3(n_i n_j \delta_{kl} + n_i n_k \delta_{jl} + n_i n_l \delta_{jk} \right. \\ &\quad \left. + n_j n_k \delta_{il} + n_j n_l \delta_{ik} + n_k n_l \delta_{ij}) - 15n_i n_j n_k n_l \right\}, \quad (2.19) \end{aligned}$$

where \vec{n} is the unit vector of $\vec{x} - \vec{x}'$, ρ is the mass density, and i, j, k, l are cartesian coordinate indices. In the rest of this paper we will use the approximation to replace $\vec{x} - \vec{x}'$ in

equation (2.19) by $\vec{x}_s - \vec{x}_{s'}$ for the pair of elementary blocks s and s' , where \vec{x}_s denotes the center of the s th block, and $\int_{V^{(s)}} \hat{T}_{ij}(\vec{x}) d^3x = \hat{T}_{ij}^{(s)}$ is the uniform non-harmonic stress tensor of the s th block.

We now combine N_0^3 elementary blocks together to form a super block with side length $N_0 L$. Given the many-body interaction in equation (2.18), the super block Hamiltonian is

$$\begin{aligned} \hat{H}_0^{\text{sup}} &= \hat{H}_0 + \hat{V}, \\ \hat{H}_0 &= \sum_{s=1}^{N_0^3} \hat{H}_0^{(s)}, \quad \hat{V} = \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}, \quad (2.20) \end{aligned}$$

where $\hat{H}_0^{(s)}$ is the non-harmonic part of Hamiltonian of the s th elementary block. We denote $\{|n^{(s)}\rangle\}$ and $\{E_n^{(s)}\}$ to be the complete and orthogonal set of eigenstates and corresponding eigenvalues of the s th elementary block Hamiltonian, $\hat{H}_0^{(s)}$. Thus $|n\rangle = \prod_s |n^{(s)}\rangle$ and $E_n = \sum_s E_n^{(s)}$ are the eigenstates and eigenvalues of $\hat{H}_0 = \sum_s \hat{H}_0^{(s)}$. We also denote $|n^{\text{sup}}\rangle$ and E_n^{sup} to be the eigenstates and eigenvalues of \hat{H}_0^{sup} . With the assumption of $\hat{V} \ll \hat{H}_0$, we can set up the relation between $|n^{\text{sup}}\rangle$, E_n^{sup} and $|n\rangle$, E_n through perturbation theory,

$$\begin{aligned} |n^{\text{sup}}\rangle &= |n\rangle + \sum_{p \neq n} \frac{\langle p | \hat{V} | n \rangle}{E_n - E_p} |p\rangle + \mathcal{O}(V^2), \\ E_n^{\text{sup}} &= E_n + \langle n | \hat{V} | n \rangle + \mathcal{O}(V^2). \quad (2.21) \end{aligned}$$

2.4. Super block Hamiltonian with external strain field

Equation (2.20) represents the Hamiltonian of super block amorphous solid without external strain field. Hereafter, we focus on the Hamiltonian with weakly imposed external strain $\mathbf{e}^{(s)}(t)$ (or equivalently, $\vec{u}^{(s)}(t)$ denotes external displacement field). It appears that only one time-dependent perturbation term $\hat{H}'_1(t) = \sum_s \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)}$ is introduced to the super block Hamiltonian. However, two extra time-dependent perturbations $\hat{H}'_2(t)$ and $\hat{H}'_3(t)$ also arise with the existence of external strain field. Here we emphasize that the energy absorption contribution of $\hat{H}'_2(t)$ and $\hat{H}'_3(t)$ are negligible compared to that of $\hat{H}'_1(t)$, and thus leave the detailed discussion of $\hat{H}'_2(t)$ and $\hat{H}'_3(t)$ in appendix B. By applying external phonon strain, the super block Hamiltonian of amorphous solid can be written as follows,

$$\begin{aligned} \hat{H}_0^{\text{sup}}(t) &= \hat{H}_0 + \hat{V} + \hat{H}'_1(t) + \hat{H}'_2(t) + \hat{H}'_3(t) \\ \hat{H}'_1(t) &= \sum_{s=1}^{N_0^3} \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)}, \quad (2.22) \end{aligned}$$

where $\hat{H}'_1(t)$, $\hat{H}'_2(t)$ and $\hat{H}'_3(t)$ are in linear order of the amplitude of external phonon wave.

Note that both of the stress tensor operator $\hat{T}_{ij}^{(s)}$ and susceptibility $\chi_{ijkl}(\omega)$ are defined from the amorphous solid block with the side length L . Starting from section 3, we will use $\hat{T}_{ij}^{(s)}(L)$ and $\chi_{ijkl}(\omega, L)$ instead of $\hat{T}_{ij}^{(s)}$ and $\chi_{ijkl}(\omega)$.

3. Resonance phonon energy absorption

As already mentioned in TTLS model, resonance phonon energy absorption rate $dE_{l,t}/dt$ is proportional to $\gamma_{l,t}^2$. The purpose of this section is to calculate resonance energy absorption with the super block Hamiltonian (recall equation (2.22)).

We first calculate energy absorption of the Hamiltonian $\hat{H}_0^{(s)}(t) = \hat{H}_0^{(s)} + \sum_{ij} e_{ij}^{(s)}(t)\hat{T}_{ij}^{(s)}$ of the elementary block with side length L . The resonance energy absorption rate of elementary block is

$$dE_\alpha(L)/dt = 2L^3 A^2 k^2 \omega (1 - e^{-\beta\omega}) \text{Im} \tilde{\chi}_\alpha(\omega, L). \quad (3.1)$$

If only considering elementary-block, we simply generalize the ratio of energy absorption from $\frac{dE_l/dt}{dE_t/dt} \propto \frac{\gamma_l^2}{\gamma_t^2}$ to $\frac{dE_l(L)/dt}{dE_t(L)/dt} \propto \frac{\text{Im} \tilde{\chi}_l(\omega, L)}{\text{Im} \tilde{\chi}_t(\omega, L)}$.

If we combine a group of elementary blocks, the many-body interaction will affect the energy absorption. We now attempt to express the ultrasonic absorption of the super block in terms of the susceptibility of the elementary blocks $\text{Im} \tilde{\chi}_\alpha(\omega, L)$. In equation (2.22) we take \hat{V} , $\hat{H}'_1(t)$, $\hat{H}'_2(t)$ and $\hat{H}'_3(t)$ as perturbations to \hat{H}_0 (for definition of \hat{H}_0 please refer to equation (2.20)). We expand the super block energy absorption up to the second order of phonon amplitude A , and to the second order of elementary block reduced susceptibility, $\text{Im} \tilde{\chi}_\alpha(\omega, L)$.

In order to calculate the energy absorption of super block glass, we use the approximation that for stress tensors in different elementary blocks, the average of the stress tensor product vanishes:

$$\langle \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \rangle_{s \neq s'} = \int_{V^{(s)}} d^3x \int_{V^{(s')}} d^3x' \sum_n \delta(E_n - E_m - \omega) \langle m | \hat{T}_{ij}(\vec{x}) | n \rangle \langle n | \hat{T}_{kl}(\vec{x}') | m \rangle_{s \neq s'} \approx 0. \quad (3.2)$$

This approximation is based on the assumption that the stress tensors (i.e. $\hat{T}_{ij}^{(s)} = \delta \hat{H}^{(s)} / \delta e_{ij}$) in different blocks are random and uncorrelated. On average these terms are much smaller than the correlation of stress tensors which belong to the same block, $\langle \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \rangle_{s=s'}$.

With the above assumption, we are now ready to calculate the energy absorption of super block Hamiltonian. Up to the second order of phonon amplitude and elementary block susceptibility, there are four contributions to the energy absorption. The corresponding mathematical details can be found in appendix C. The first term $dE_\alpha^{(0)}(L)/dt$ comes from the second order perturbation of $\hat{H}'_1(t)$. The result is simply the summation of elementary block energy absorption,

$$dE_\alpha^{(0)}(L)/dt = N_0^3 dE_\alpha(L)/dt. \quad (3.3)$$

The second term $dE_\alpha^{(1)}(L)/dt$ comes from the second order perturbation of $\hat{H}'_1(t)$ as well. It is also the first order perturbation of many-body interaction \hat{V} :

$$dE_\alpha^{(1)}(L)/dt = (1 - e^{-\beta\omega}) \frac{4N_0^3 L^3 A^2 k^2 \ln N_0}{\rho c_\alpha^2} \omega \text{Im} \tilde{\chi}_\alpha(\omega, L) \text{Re} \tilde{\chi}_\alpha(\omega, L), \quad (3.4)$$

where

$$\text{Re} \tilde{\chi}_\alpha(\omega, L) = \frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\Omega \text{Im} \chi_\alpha(\Omega, L) d\Omega}{(1 - e^{-\beta\omega})(\Omega^2 - \omega^2)} \quad (3.5)$$

is the ‘real part of the reduced non-harmonic susceptibility’ obtained through Kramers–Kronig relation. Similarly, the third term $dE_\alpha^{(2)}(L)/dt$ and the fourth term $dE_\alpha^{(3)}(L)/dt$ are derived from the second order perturbation of $\hat{H}'_2(t)$ and $\hat{H}'_3(t)$, respectively. In appendix B we provide detailed calculations of $dE_\alpha^{(2)}(L)/dt$ and $dE_\alpha^{(3)}(L)/dt$, and prove that $dE_\alpha^{(2,3)}(L)/dt \ll dE_\alpha^{(0,1)}(L)/dt$. Finally, we neglect $dE_\alpha^{(2)}(L)/dt$ and $dE_\alpha^{(3)}(L)/dt$ to obtain the energy absorption of super block glass,

$$dE_\alpha^{\text{sup}}(L)/dt = dE_\alpha^{(0)}(L)/dt + dE_\alpha^{(1)}(L)/dt. \quad (3.6)$$

On the other hand, we notice that $dE_\alpha(N_0L)/dt$ can be expressed in terms of $\text{Im} \tilde{\chi}_\alpha(\omega, N_0L)$ which is the susceptibility of super block,

$$dE_\alpha(N_0L)/dt = 2(N_0L)^3 A^2 k^2 \omega (1 - e^{-\beta\omega}) \text{Im} \tilde{\chi}_\alpha(\omega, N_0L). \quad (3.7)$$

We notice that $dE_\alpha(N_0L)/dt = dE_\alpha^{\text{sup}}(L)/dt$. Since the equal signs hold in equations (3.6) and (3.7), we can now construct the renormalization equation of susceptibility in the following section.

4. Real space renormalization of resonance energy absorption

In this section, we perform real space renormalization technique to carry out the recursion relation of resonance phonon energy absorption between elementary and super block amorphous solid. To this end, we combine N_0^3 elementary blocks with the side length L to form a super block with the side length N_0L . Similarly, we treat this super block as a new elementary block, and combine N_0^3 new elementary blocks to obtain a new super block of amorphous solid with the side length N_0^2L . We repeat this renormalization process initiating from unit blocks at microscopic length scale L_1 to macroscopic blocks at experimental length scale R . In the process of renormalization, the elementary block energy absorption in $(n+1)$ th step is equivalent to super block energy absorption in n th step. This relation is mathematically formulated by equations (3.6) and (3.7), which we use to construct the renormalization equation of $\text{Im} \tilde{\chi}_\alpha(\omega, L_n)$,

$$\text{Im} \tilde{\chi}_\alpha(\omega, L_{n+1}) = \text{Im} \tilde{\chi}_\alpha(\omega, L_n) + \frac{2 \ln N_0}{\rho c_\alpha^2} K_\alpha(\omega, L_n) [\text{Im} \tilde{\chi}_\alpha(\omega, L_n)]^2, \quad (4.1)$$

where $L_{n+1} = N_0L_n$ is the side length of step $n+1$ renormalization procedure, and

$$K_\alpha(\omega, L_n) \stackrel{\text{def}}{=} \text{Re} \tilde{\chi}_\alpha(\omega, L_n) / \text{Im} \tilde{\chi}_\alpha(\omega, L_n) \quad (4.2)$$

is the ratio between the real part and the imaginary part of reduced resonance susceptibility. As we repeatedly employ this renormalization scheme, it will bring out non-harmonic susceptibilities at experimental length scale R .

Table 1. Theory versus experiment of γ_l/γ_t (experimental data summarized by Meissner and Berret [1]).

Material	γ_l (eV)	γ_t (eV)	$(\gamma_l/\gamma_t)_{\text{exp}}$	c_l (km s ⁻¹)	c_t (km s ⁻¹)	$(\gamma_l/\gamma_t)_{\text{theo}} = c_l/c_t$	$\frac{(\text{theo-exp})}{\text{exp}}$
a-SiO ₂	1.04	0.65	1.60	5.80	3.80	1.53	-4.38%
BK7	0.96	0.65	1.48	6.20	3.80	1.63	+10.1%
As ₂ S ₃	0.26	0.17	1.53	2.70	1.46	1.85	+20.9% (measured from K_3)
LaSF-7	1.46	0.92	1.59	5.64	3.60	1.57	-1.26% (measured from K_3)
SF4	0.72	0.48	1.50	3.78	2.24	1.69	+12.7%
SF59	0.77	0.49	1.57	3.32	1.92	1.73	+10.2%
V52	0.87	0.52	1.67	4.15	2.25	1.84	+10.4%
BALNA	0.75	0.45	1.67	4.30	2.30	1.87	+12.0%
LAT	1.13	0.65	1.74	4.78	2.80	1.71	-1.72%
a-Se	0.25	0.14	1.79	2.00	1.05	1.90	+6.14%
Zn-Glass	0.70	0.38	1.84	4.60	2.30	2.00	+8.70%
PMMA [31]	0.39	0.27	1.44	3.15	1.57	2.01	+39.6% (large deviation)
PS	0.20	0.13	1.54	2.80	1.50	1.87	+21.4% (measured from K_3)
LiCl-7H ₂ O	0.62	0.39	1.59	4.00			
PC	0.28	0.18	1.56	2.97			
Epoxy	0.35	0.22	1.59	3.25			

We now have a further look at equations (4.1) and (4.2). The evolve of $\text{Im } \tilde{\chi}_\alpha(\omega, L_n)$ is dominated by the sign of $K_\alpha(\omega, L_n)$. When $K_\alpha(\omega, L_n) > 0$, the susceptibility diverges as $R \rightarrow \infty$, which means there is no fixed point of $\text{Im } \tilde{\chi}_\alpha(\omega, R)$. When $K_\alpha(\omega, L_n) < 0$, $\text{Im } \tilde{\chi}_\alpha(\omega, L_n)$ logarithmically decreases as $R \rightarrow \infty$, which means it converges to a fixed point. From equation (2.16), given that non-harmonic susceptibility $\text{Im } \tilde{\chi}_\alpha(\omega, L_n)$ is positive, $K_\alpha(\omega, L_n)$ is positive (negative) when $\text{Re } \tilde{\chi}_\alpha(\omega, L_n) > 0$ (< 0). Hence, it is essential to figure out the sign of $\text{Re } \tilde{\chi}_\alpha(\omega, L_n)$.

If we integrate equation (3.5) directly, we are yet unable to quantitatively calculate $\text{Re } \tilde{\chi}_\alpha(\omega, L_n)$ and further determine the sign of it due to the general form of $\text{Im } \tilde{\chi}_\alpha(\omega, L_n)$. However, within ultrasonic experiments we argue that the sign of $\text{Re } \tilde{\chi}_\alpha(\omega, L_n)$ is the same as that of space-averaged TTLS susceptibility given by equation (2.10). Consequently, $K_\alpha(\omega, L_n)$ becomes negative as well and $\text{Im } \tilde{\chi}_\alpha(\omega, L_n)$ logarithmically decreases with the increase of R . Furthermore, this result is in agreement with the small and universal internal friction Q^{-1} in the ultrasonic experiments by Pohl *et al* [12], and the small and universal slope of logarithmic temperature dependence of sound velocity shift [13, 14, 16, 18–23] $\Delta c_\alpha/c_\alpha$.

The above argument is proposed when $\beta\hbar\omega \ll 1$, which is fulfilled within ultrasonic conditions [1, 24, 25] when $10 \text{ MHz} \lesssim f \lesssim 1 \text{ GHz}$ and $0.1 \text{ K} \lesssim T \lesssim 17 \text{ K}$. We expect the universal behavior to fail if f and T violate $\beta\hbar\omega \ll 1$. Since $\text{Re } \chi_\alpha(\omega)|_{\text{TTLS}} < 0$ is no longer valid (see equation (2.10)), we can expect that the sign of $\text{Re } \tilde{\chi}_\alpha(\omega, L_n)$ is the same as that of $\text{Re } \chi_\alpha(\omega)|_{\text{TTLS}}$ which is positive. The imaginary part of susceptibility $\text{Im } \tilde{\chi}_\alpha(\omega, L_n)$ diverges and there is no fixed point. Consequently, the thermal and acoustic properties of low-temperature amorphous solids are no longer universal, including the γ_l/γ_t ratio.

Next, we denote $K_\alpha(\omega, L_n) = -|K_\alpha(\omega, L_n)|$. We also define ‘the change of non-harmonic susceptibility’ as $\Delta \text{Im } \tilde{\chi}_\alpha(\omega, L_n) = \text{Im } \tilde{\chi}_\alpha(\omega, L_{n+1}) - \text{Im } \tilde{\chi}_\alpha(\omega, L_n)$, which is much smaller than the non-harmonic susceptibility, i.e.

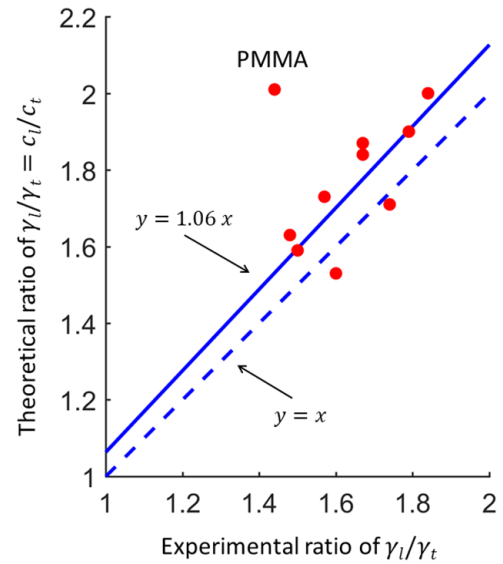


Figure 1. The least square fitting for the ratio γ_l/γ_t between the theory and the experiment. The least square fitting is $y = 1.06x$ for the data of nine materials excluding PMMA. The linear correlation coefficient is $r = 0.803$. The dashed line is our original theoretical prediction $(\gamma_l/\gamma_t)_{\text{theo}} = c_l/c_t$.

$\Delta \text{Im } \tilde{\chi}_\alpha(\omega, L_n) \ll \text{Im } \tilde{\chi}_\alpha(\omega, L_n)$. We repeat the renormalization process from microscopic length scale L_1 to experimental length scale R to obtain non-harmonic susceptibility as follows,

$$\frac{1}{\text{Im } \tilde{\chi}_\alpha(\omega, R)} = \frac{1}{\text{Im } \tilde{\chi}_\alpha(\omega, L_1)} + \frac{2 \ln N_0}{\rho c_\alpha^2} \sum_{n=1}^{\log_{N_0} \frac{R}{L_1}} |K_\alpha(\omega, L_n)|. \quad (4.3)$$

To compare the first and second terms of equation (4.3), we argue that the second term is roughly $\frac{2}{\rho c_\alpha^2} \ln \left(\frac{R}{L_1} \right) |K_\alpha(\omega)|$. As stated by Vural and Leggett [2], the experimental length

scale is the phonon wavelength $R = 2\pi c_\alpha/\omega$. The input phonon frequency is of order of $f \sim 10^6$ Hz with the wavelength $R \sim 10^{-3}$ m in the ultrasonic measurements from Meissner and Berret [1]. The number of renormalization steps, $\log_{N_0}(R/L_1)$, logarithmically depends on the microscopic length scale L_1 , meaning that the number of steps is not sensitive to the choice of L_1 . We therefore choose $L_1 \sim 50$ Å suggested by Vural and Leggett [2]. The factor $\ln(R/L_1)$ is of order 20, yielding the second term of equation (4.3) much greater than the first. Hence, $\text{Im } \tilde{\chi}_\alpha(\omega, R)$ is dominated by the second term, with

$$\text{Im } \tilde{\chi}_\alpha(\omega, R) = \frac{\rho c_\alpha^2}{2 \ln N_0} \left(\sum_{n=1}^{\log_{N_0}(R/L_1)} |K_\alpha(\omega, L_n)| \right)^{-1}. \quad (4.4)$$

The ratio between longitudinal and transverse energy absorption is

$$\frac{dE_l(R)/dt}{dE_t(R)/dt} = \frac{\text{Im } \tilde{\chi}_l(\omega, R)}{\text{Im } \tilde{\chi}_t(\omega, R)} = \frac{c_l^2 \sum_n |K_l(\omega, L_n)|}{c_t^2 \sum_n |K_t(\omega, L_n)|}. \quad (4.5)$$

To further simplify equation (4.5), let us rewind the assumption we made at the end of section 2.2. As stated by equation (2.8), the frequency dependence of longitudinal TTLS susceptibility is the same as that of transverse TTLS susceptibility, i.e. they are both proportional to $\tanh(\beta\hbar\omega/2)$. Likewise, we assume that the frequency dependences of $\text{Im } \tilde{\chi}_l(\omega, L_n)$ and $\text{Im } \tilde{\chi}_t(\omega, L_n)$ are roughly the same. Since $\text{Re } \tilde{\chi}_\alpha(\omega, L_n)$ is obtained from $\text{Im } \tilde{\chi}_\alpha(\omega, L_n)$ through Kramers–Kronig relation (see equation (3.5)), $K_\alpha(\omega, L_n) = \text{Re } \tilde{\chi}_\alpha(\omega, L_n)/\text{Im } \tilde{\chi}_\alpha(\omega, L_n)$ is roughly the same for longitudinal and transverse phonons, i.e. $K_l(\omega, L_n) \approx K_t(\omega, L_n)$. Under this assumption, the ratio of resonance energy absorption is

$$\frac{dE_l(R)/dt}{dE_t(R)/dt} = \frac{\text{Im } \tilde{\chi}_l(\omega, R)}{\text{Im } \tilde{\chi}_t(\omega, R)} = \frac{c_l^2}{c_t^2}. \quad (4.6)$$

The ratio of longitudinal and transverse real part of susceptibility is calculated through Kramers–Kronig relation, with

$$\frac{\text{Re } \tilde{\chi}_l(\omega, R)}{\text{Re } \tilde{\chi}_t(\omega, R)} = \frac{c_l^2}{c_t^2}. \quad (4.7)$$

To summarize, the ratio of imaginary part of non-harmonic susceptibilities ultimately converges to a fixed point, given by equation (4.6). It gives the theoretical result of our model, $\gamma_l/\gamma_t = c_l/c_t$. This is an experimentally measurable quantity containing no adjustable parameters or other microscopic unmeasurable quantities [26–30]. It paves the way for future experimental investigation to deeper understand the mechanism of low-energy excitations of amorphous solids.

5. Theory versus experimental data

We are now able to compare our theoretical result, i.e. $\gamma_l/\gamma_t = c_l/c_t$, with the experimental data of 16 different non-metallic amorphous materials [1]. In table 1, we use ‘ $(\gamma_l/\gamma_t)_{\text{exp}}$ ’ to stand for the experimental data, and ‘ $(\gamma_l/\gamma_t)_{\text{theo}} = c_l/c_t$ ’ to represent our theoretical result.

Among the 16 amorphous materials [1], LiCl-7H₂O, PC and Epoxy lack the data of transverse sound velocity. For the rest of 13 amorphous materials, we discard the data of PS, As₂S₃ and LASF-7 due to the reason that the experimental measurements of γ_l/γ_t were not obtained directly from sound velocity shift measurements in resonance regime [1]. They were obtained from K_3 , the coefficient of the relaxation procedure which is different from the resonance procedure and relies on TTLS model. As expected, the experimental results of nine materials agree reasonably well with our theory, except PMMA is the only outlier which deviates from the theory by 40%. Least square method is used to examine the statistical significance between theory and experiment. For the nine materials excluding PMMA, the linear fitting is $(\gamma_l/\gamma_t)_{\text{theo}} = 1.06 (\gamma_l/\gamma_t)_{\text{exp}}$, with the linear correlation coefficient $r = 0.803$. We plot the relationship between theory and experiment in figure 1, where the x -axis and the y -axis stand for the experimental and theoretical ratio γ_l/γ_t , respectively. The above results demonstrate that our theory properly explains and validates the universal ratio of γ_l/γ_t for these nine materials.

Additionally, we discuss the outlier of material PMMA. According to Lin and Lee [32], the acoustic coefficients of PMMA from different literatures vary for more than the factor of 4, which means that the ultrasonic measurements on this material tend to fluctuate a lot. This could lead to the large deviation between theory and experiment of PMMA.

6. Summary and outlook

In this paper we have studied the universal ratio of TTLS-phonon coupling constants of amorphous solids at low temperatures. By using the model proposed by Vural and Leggett, we apply real space renormalization technique to prove that γ_l/γ_t flows to c_l/c_t , the ratio of sound velocities. This is an experimentally measurable quantity with no adjustable parameters.

To achieve that goal, we first separate the Hamiltonian of an elementary block of amorphous solid into two parts, the purely elastic phonon excitations, and the leftover part, namely non-harmonic Hamiltonian. We expand the non-harmonic Hamiltonian in orders of long wavelength phonon strain field. The first order expansion is the coupling between phonon strain and non-harmonic stress tensor operators. The zeroth-order expansion of non-harmonic Hamiltonian and the non-harmonic stress tensor are therefore the multiple-level-system generalization of TTLS model. We then combine elementary blocks together to form a super block of amorphous solid. By allowing the exchange of virtual phonons, we obtain many-body interaction between stress tensors, which is the mutual interaction between elementary blocks.

Furthermore, by calculating resonance phonon energy absorption of elementary and super block Hamiltonians, we set up the recursion relation of energy absorption between small and large length scale amorphous solids, which is the real space renormalization equation. As the length scale increases, the ratio between longitudinal and transverse phonon energy absorption is determined by the mutual interaction between

elementary blocks. It flows to $(dE_l/dt)/(dE_t/dt) = c_l^2/c_t^2$ at experimental length scale.

Among 16 amorphous materials summarized by Meissner and Berret [1], we discard the data of LiCl-7H₂O, PC and Epoxy due to the absence of transverse sound velocity data. In addition, we discard the data of PS, As₂S₃ and LASF-7, since the measurement of γ_l/γ_t is obtained from K_3 , the coefficient of the relaxation procedure which relies on the TTLS model and is therefore different from resonance energy absorption process. In this paper, we state that the data of nine materials are in line with our theory. The only outlier PMMA deviating from our theory could possibly be caused by large fluctuations of ultrasound measurements in different literatures. We hope this paper will draw the attention of experimentalists and more experimental data of γ_l/γ_t in future to verify our theory.

Besides the universal TTLS-phonon coupling ratio, a number of other works [30, 33–35, 36–38] also approach dielectric and acoustic problems by considering this long-range interaction. For example, Burin [33] analyzed the influence of $1/r^3$ long-range interaction of two-level-systems on AC dielectric susceptibility in SiO_x. This interaction leads to the ‘dipole gap’, which is a significant decrease in the low-energy density of states and further reflected from the drastic change of dielectric susceptibility. The instantaneous increase in AC dielectric susceptibility and the consequent observed logarithmic relaxation [36] are qualitatively consistent with the model of weakly interacted two-level-systems, whereas the isolated two-level-systems are not adequate to explain them. Generalized from the previous theory [33] and the experiment [36], Nalbach *et al* explored the non-equilibrium dielectric susceptibility of polyester glass Mylar and borosilicate glass BK7 with temporary electric field [34]. Inspired by the dipole gap theory, the authors pointed out that the observed excess dielectric response originates from the non-equilibrium dynamics of coupled clusters of two-level-systems.

Despite the fact that this paper considers mechanical susceptibility rather than dielectric susceptibility, the form of long-range interactions are similar, and stress tensor operators are simply generalized from two-level-systems. It is therefore interesting to ask that given a strong mechanical strain to a glass sample, whether a similar increase of mechanical susceptibility and the subsequent logarithmic relaxation can be observed. It is on our agenda to explore the mechanical analog of dielectric relaxation in low-temperature amorphous solids.

The physical nature of two-level-systems has been a long-standing question. Previous literatures [5, 6] suggested that two-level-systems could possibly stem from a group of atoms sitting in two equilibrium positions. Later, inspired by off-center and rotational impurities in disordered lattices and amorphous materials, Schechter and Stamp [30] proposed a different picture which includes two types of two-level-systems. One is asymmetric under inversion and thus interacts strongly with strain field, while the other one is inversion symmetric and interacts weakly with strain. Both of them generate $1/r^3$ long-range interactions, but the interaction between inversion asymmetric two-level-systems is far greater than that between symmetric ones. Below a critical temperature the asymmetric two-level-systems are frozen and the active

symmetric ones are weakly-interacting pseudo-spins. The authors addressed that universal properties appear below the critical temperature due to these symmetric two-level-systems. However at higher temperatures, the Hamiltonian involves only the inversion asymmetric two-level-systems, which could possibly be related to glass transition problems [35]. The model proposed by Schechter and Stamp attempts to link the seemingly unrelated two features of glasses, known as low-temperature quantum universalities and high-temperature glass transition behaviors.

Inspired by these works [30, 35], we ask if the stress tensor operators could be separated into inversion symmetric and asymmetric parts. At low-temperatures only weakly-interacting symmetric stress tensors are active. This is in accordance with our perturbative renormalization process that the many-body interaction is weak compared to elementary block Hamiltonian. However, this weak interaction is not negligible in the process of renormalization. A marginally irrelevant behavior of mechanical susceptibility is obtained through this procedure. The idea of inversion symmetric and asymmetric stress tensors could open the door to a deeper understanding of low-temperature universalities, as well as the connection to high-temperature glass transition problems, which will be the next step of our research.

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Appendix A. Deriving non-harmonic stress–stress many-body interaction between elementary blocks

The non-harmonic stress–stress many-body interaction \hat{V} and the coefficient $\Lambda_{ijkl}^{(ss')}|_{\text{JL}}$ was originally derived by Joffrin and Levelut [3]. We denote them as $\hat{V}|_{\text{JL}}$ and $\Lambda_{ijkl}^{(ss')}|_{\text{JL}}$.

$$\begin{aligned}\hat{V}|_{\text{JL}} &= \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \Lambda_{ijkl}^{(ss')}|_{\text{JL}} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \\ \Lambda_{ijkl}^{(ss')}|_{\text{JL}} &= -\frac{\tilde{\Lambda}_{ijkl}(\vec{n})|_{\text{JL}}}{8\pi\rho c_l^2 |\vec{x}_s - \vec{x}_s'|^3} \\ \tilde{\Lambda}_{ijkl}(\vec{n})|_{\text{JL}} &= -2(\delta_{jl} - 3n_j n_l) \delta_{ik} \\ &+ 2\alpha \{ -(\delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{jk} \delta_{il}) + 3(n_i n_j \delta_{kl} + n_i n_k \delta_{jl} \\ &+ n_i n_l \delta_{jk} + n_j n_k \delta_{il} + n_j n_l \delta_{ik} + n_k n_l \delta_{ij}) - 15n_i n_j n_k n_l \},\end{aligned}\quad (\text{A.1})$$

where $\alpha = 1 - c_l^2/c_t^2$, i, j, k are cartesian indices, and \vec{n} is the unit vector of $\vec{x}_s - \vec{x}_s'$.

Next, we perform our derivation of \hat{V} by considering the Hamiltonian of super block formed by a group of elementary blocks. The Hamiltonian is the summation of long wavelength

phonon Hamiltonian, the coupling between the stress tensors and phonon strain field. The zeroth order non-harmonic Hamiltonian is given as

$$\hat{H}^{\text{tot}} = \sum_{\vec{q}, \mu=l,t} \left(\frac{|p_\mu(\vec{q})|^2}{2m} + \frac{1}{2} m \omega_{\vec{q}\mu}^2 |u_\mu(\vec{q})|^2 \right) + \sum_s \sum_{ij} e_{ij}^{(s)} \hat{T}_{ij}^{(s)} + \hat{H}_0^{\text{non}}, \quad (\text{A.2})$$

where μ is the phonon polarization, m is the mass of elementary block, $s = 1, 2, \dots, N_0^3$ labels these N_0^3 elementary blocks, $p_\mu(\vec{q})$ and $u_\mu(\vec{q})$ are momentum and displacement operators of phonon modes, and $e_{ij}^{(s)}$ is phonon strain field. We Fourier transform the phonon displacement field $\vec{u}^{(s)}$ to momentum space $\vec{u}_\mu(\vec{q})$:

$$u_i^{(s)} = \frac{1}{\sqrt{N}} \sum_{\vec{q}, \mu=l,t} u_\mu(\vec{q}) \mathbf{e}_{\mu i}(\vec{q}) e^{i\vec{q} \cdot \vec{x}_s}, \quad (\text{A.3})$$

where $\mathbf{e}_\mu(\vec{q})$ is the unit vector of phonon vibration direction, and N is the number density of elementary block. We denote L as the side length of elementary block, so we have $Nm/L^3 = \rho$, with ρ representing the mass density. For longitudinal phonon mode, $\mu = l$ and

$$\vec{e}_l(\vec{q}) = \vec{q}/q, \quad (\text{A.4})$$

whereas for transverse phonon mode $\mu = t_1, t_2$,

$$\begin{aligned} \vec{e}_{t_1}(\vec{q}) \cdot \vec{q} = \vec{e}_{t_2}(\vec{q}) \cdot \vec{q} = \vec{e}_{t_1}(\vec{q}) \cdot \vec{e}_{t_2}(\vec{q}) = 0 \\ \sum_{\mu=t_1, t_2} \mathbf{e}_{\mu i}(\vec{q}) \mathbf{e}_{\mu j}(\vec{q}) = \delta_{ij} - \frac{q_i q_j}{q^2}. \end{aligned} \quad (\text{A.5})$$

The strain field $e_{ij}^{(s)}$ in terms of momentum-space phonon displacement $u_\mu(\vec{q})$ is therefore expressed as

$$e_{ij}^{(s)} = \frac{1}{2\sqrt{N}} \sum_{\vec{q}, \mu} i u_\mu(\vec{q}) e^{i\vec{q} \cdot \vec{x}_s} [q_j \mathbf{e}_{\mu i}(\vec{q}) + q_i \mathbf{e}_{\mu j}(\vec{q})]. \quad (\text{A.6})$$

Since $u_i^{(s)}$ is real, we have $u_\mu(\vec{q}) = u_\mu^*(-\vec{q})$. By substituting the identity $\sum_{\vec{q}} f(\vec{q}) = \sum_{\vec{q}} \frac{1}{2} [f(\vec{q}) + f(-\vec{q})]$ we obtain the stress-strain coupling term as follows,

$$\begin{aligned} \sum_s \sum_{ij} e_{ij}^{(s)} \hat{T}_{ij}^{(s)} = \frac{1}{4\sqrt{N}} \sum_{ij} \sum_s \sum_{\vec{q}, \mu=l,t} \left[\left(i u_\mu(\vec{q}) e^{i\vec{q} \cdot \vec{x}_s} \right) \right. \\ \left. + \left(i u_\mu(\vec{q}) e^{i\vec{q} \cdot \vec{x}_s} \right)^* \right] [q_j \mathbf{e}_{\mu i}(\vec{q}) + q_i \mathbf{e}_{\mu j}(\vec{q})] \hat{T}_{ij}^{(s)}. \end{aligned} \quad (\text{A.7})$$

The stress-strain coupling is linear in $u_\mu(\vec{q})$. We therefore combine it with phonon Hamiltonian which is quadratic in $u_\mu(\vec{q})$ and ‘complete the square’. An extra term ‘ $-m\omega_{\vec{q}\mu}^2 |u_\mu^{(0)}(\vec{q})|^2/2$ ’ is left after completing the square:

$$\hat{H}^{\text{tot}} = \sum_{\vec{q}, \mu=l,t} \left(\frac{|p_\mu(\vec{q})|^2}{2m} + \frac{m\omega_{\vec{q}\mu}^2}{2} |u_\mu(\vec{q}) - u_\mu^{(0)}(\vec{q})|^2 - \frac{m\omega_{\vec{q}\mu}^2}{2} |u_\mu^{(0)}(\vec{q})|^2 \right) + \hat{H}_0^{\text{non}}, \quad (\text{A.8})$$

where the ‘equilibrium position’ $u_\mu^{(0)}(\vec{q})$ in equation (A.8) is given by

$$u_\mu^{(0)}(\vec{q}) = \frac{i}{2\sqrt{N}m\omega_{\vec{q}\mu}^2} \sum_{ij} \sum_s [q_j \mathbf{e}_{\mu i}(\vec{q}) + q_i \mathbf{e}_{\mu j}(\vec{q})] \hat{T}_{ij}^{(s)} e^{-i\vec{q} \cdot \vec{x}_s}. \quad (\text{A.9})$$

Now we calculate this extra term $-m\omega_{\vec{q}\mu}^2 |u_\mu^{(0)}(\vec{q})|^2/2$ in detail, which contains the many-body interaction between stress tensors. We split it into two parts \hat{U} and \hat{V} (see equation (A.10) below). \hat{U} is the interaction between stress tensors in the same elementary block, and \hat{V} is the interaction between stress tensors in different elementary blocks:

$$\begin{aligned} - \sum_{\vec{q}, \mu} \frac{m\omega_{\vec{q}\mu}^2}{2} |u_\mu^{(0)}(\vec{q})|^2 = \hat{U} + \hat{V} \\ \hat{U} = - \sum_{\vec{q}, \mu} \frac{1}{8Nm\omega_{\vec{q}\mu}^2} \sum_{ijkl} [q_j \mathbf{e}_{\mu i}(\vec{q}) + q_i \mathbf{e}_{\mu j}(\vec{q})] [q_k \mathbf{e}_{\mu l}(\vec{q}) \\ + q_l \mathbf{e}_{\mu k}(\vec{q})] \sum_s \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s)} \\ \hat{V} = - \sum_{\vec{q}, \mu} \frac{1}{8Nm\omega_{\vec{q}\mu}^2} \sum_{ijkl} [q_j \mathbf{e}_{\mu i}(\vec{q}) + q_i \mathbf{e}_{\mu j}(\vec{q})] [q_k \mathbf{e}_{\mu l}(\vec{q}) \\ + q_l \mathbf{e}_{\mu k}(\vec{q})] \sum_{s \neq s'} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \cos(\vec{q} \cdot (\vec{x}_s - \vec{x}_{s'})). \end{aligned} \quad (\text{A.10})$$

Since \hat{V} stands for the many-body interaction between different elementary blocks, the number of interactions quadratically increases as the number of elementary blocks increases. We hereby name \hat{V} as non-harmonic stress–stress many-body interaction. It can be further simplified as follows,

$$\begin{aligned} \hat{V} = \sum_{s \neq s'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \\ \Lambda_{ijkl}^{(ss')} = \frac{1}{2\rho c_t^2} \sum_{\vec{q}, \frac{2\pi}{L_{\text{max}}} \leq |\vec{q}| \leq \frac{2\pi}{L}} \left[\alpha \left(\frac{q_i q_j q_k q_l}{q^4} \right) \right. \\ \left. - \frac{1}{4} \left(\frac{q_j q_l \delta_{ik} + q_j q_k \delta_{il} + q_i q_l \delta_{jk} + q_i q_k \delta_{jl}}{q^2} \right) \right] \cos(\vec{q} \cdot \vec{x}_{ss'}), \end{aligned} \quad (\text{A.11})$$

where $\vec{x}_{ss'} = \vec{x}_s - \vec{x}_{s'}$, and L_{max} stands for the side length of the super block. In the main text we combine N_0^3 elementary blocks to form a super block with side length $L_{\text{max}} = N_0 L$. We use the identities $\sum_{\vec{q}} (q_i q_j q_k q_l / q^4) e^{i\vec{q} \cdot \vec{x}_{ss'}} = \sum_{\vec{q}} (q_i q_j q_k q_l / q^4) e^{-i\vec{q} \cdot \vec{x}_{ss'}}$ and $\sum_{\vec{q}} ((q_j q_l \delta_{ik} + q_j q_k \delta_{il} + q_i q_l \delta_{jk} + q_i q_k \delta_{jl}) / q^2) e^{i\vec{q} \cdot \vec{x}_{ss'}} = \sum_{\vec{q}} ((q_j q_l \delta_{ik} + q_j q_k \delta_{il} + q_i q_l \delta_{jk} + q_i q_k \delta_{jl}) / q^2) e^{-i\vec{q} \cdot \vec{x}_{ss'}}$ to rewrite equation (A.11) as follows,

$$\begin{aligned} \hat{V} = \sum_{s \neq s'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \\ \Lambda_{ijkl}^{(ss')} = \frac{1}{2\rho c_t^2} \sum_{\vec{q}, \frac{2\pi}{L_{\text{max}}} \leq |\vec{q}| \leq \frac{2\pi}{L}} \left[\alpha \left(\frac{q_i q_j q_k q_l}{q^4} \right) \right. \\ \left. - \frac{1}{4} \left(\frac{q_j q_l \delta_{ik} + q_j q_k \delta_{il} + q_i q_l \delta_{jk} + q_i q_k \delta_{jl}}{q^2} \right) \right] e^{i\vec{q} \cdot \vec{x}_{ss'}}. \end{aligned} \quad (\text{A.12})$$

Before making further progresses and approximations, we emphasize that equation (A.12) are the most original definition of $\Lambda_{ijkl}^{(ss')}$.

We then apply approximations to equation (A.12) to reduce the form of $\Lambda_{ijkl}^{(ss')}$. We notice that in the integration domain we are interested in, the phonon wave length λ is always greater

than the side length L of elementary block. We therefore take the approximation to replace $\sum_{\vec{q}}$ with the continuum limit $\int \frac{d^3q}{(2\pi)^3}$. Equation (A.12) is simplified as

$$\Lambda_{ijkl}^{(ss')} = \frac{1}{2\rho c_t^2} \int \frac{d^3q}{(2\pi)^3} \left[\alpha \left(\frac{q_i q_j q_k q_l}{q^4} \right) - \frac{1}{4} \left(\frac{q_j q_l \delta_{ik} + q_j q_k \delta_{il} + q_i q_l \delta_{jk} + q_i q_k \delta_{jl}}{q^2} \right) \right] e^{i\vec{q} \cdot \vec{x}_{ss'}}. \quad (\text{A.13})$$

To evaluate equation (A.13) we consider the following two integrals

$$f_{ijkl}^{(1)} = \int \frac{d^3q}{(2\pi)^3} \frac{q_i q_j q_k q_l}{q^4} e^{i\vec{q} \cdot \vec{x}} \quad f_{jl}^{(2)} = \int \frac{d^3q}{(2\pi)^3} \frac{q_j q_l}{q^2} e^{i\vec{q} \cdot \vec{x}}. \quad (\text{A.14})$$

Here we introduce a new parameter λ into the denominator $1/q^2$, and calculate $f_{ijkl}^{(1)}(\lambda)$ and $f_{jl}^{(2)}(\lambda)$:

$$f_{ijkl}^{(1)}(\lambda) = \left(\frac{\partial}{\partial x_i} \frac{\partial}{\partial x_j} \frac{\partial}{\partial x_k} \frac{\partial}{\partial x_l} \right) \int \frac{d^3q}{(2\pi)^3} \frac{1}{(q^2 + \lambda^2)^2} e^{i\vec{q} \cdot \vec{x}} \\ f_{jl}^{(2)}(\lambda) = - \left(\frac{\partial}{\partial x_j} \frac{\partial}{\partial x_l} \right) \int \frac{d^3q}{(2\pi)^3} \frac{1}{(q^2 + \lambda^2)} e^{i\vec{q} \cdot \vec{x}}. \quad (\text{A.15})$$

By using contour integral and choosing the pole at $q = -i\lambda$, we have,

$$f_{ijkl}^{(1)}(\lambda) = \left(\frac{\partial}{\partial x_i} \frac{\partial}{\partial x_j} \frac{\partial}{\partial x_k} \frac{\partial}{\partial x_l} \right) \frac{1}{8\pi\lambda} e^{-\lambda x} \\ f_{jl}^{(2)}(\lambda) = - \left(\frac{\partial}{\partial x_j} \frac{\partial}{\partial x_l} \right) \frac{1}{4\pi x} e^{-\lambda x}. \quad (\text{A.16})$$

Taking the limit $\lambda \rightarrow 0$ finally leads to the following results,

$$\lim_{\lambda \rightarrow 0} f_{ijkl}^{(1)}(\lambda) = \frac{1}{8\pi x^3} \left\{ (\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{jk}\delta_{il}) - 3(n_i n_j \delta_{kl} + n_i n_k \delta_{jl} + n_i n_l \delta_{jk} + n_j n_k \delta_{il} + n_j n_l \delta_{ik} + n_k n_l \delta_{ij}) + 15n_i n_j n_k n_l \right\} \\ \lim_{\lambda \rightarrow 0} f_{jl}^{(2)}(\lambda) = \frac{1}{4\pi x^3} (\delta_{jl} - 3n_j n_l). \quad (\text{A.17})$$

Plugging the above results into \hat{V} yields the coefficient $\Lambda_{ijkl}^{(ss')}$ of the many-body interaction as follows,

$$\hat{V} = \sum_{s \neq s'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \\ \Lambda_{ijkl}^{(ss')} = - \frac{\tilde{\Lambda}_{ijkl}(\vec{n})}{8\pi \rho c_t^2 |x_s - x'_s|^3} \\ \tilde{\Lambda}_{ijkl}(\vec{n}) = \frac{1}{4} \{ (\delta_{jl} - 3n_j n_l) \delta_{ik} + (\delta_{jk} - 3n_j n_k) \delta_{il} \\ + (\delta_{ik} - 3n_i n_k) \delta_{jl} + (\delta_{il} - 3n_i n_l) \delta_{jk} \} \\ + \frac{1}{2} \alpha \{ -(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{jk}\delta_{il}) + 3(n_i n_j \delta_{kl} \\ + n_i n_k \delta_{jl} + n_i n_l \delta_{jk} + n_j n_k \delta_{il} + n_j n_l \delta_{ik} + n_k n_l \delta_{ij}) - 15n_i n_j n_k n_l \}. \quad (\text{A.18})$$

Now let us compare $\Lambda_{ijkl}^{(ss')}$ in equation (A.18) with $\Lambda_{ijkl}^{(ss')}|_{\text{JL}}$ in equation (A.1). There are three differences between our

result and the result derived by Joffrin and Levelut. To illustrate these differences, we separate $\tilde{\Lambda}_{ijkl}(\vec{n}) = \tilde{\Lambda}_{ijkl}^{(1)}(\vec{n}) + \tilde{\Lambda}_{ijkl}^{(2)}(\vec{n})$ and $\tilde{\Lambda}_{ijkl}(\vec{n})|_{\text{JL}} = \tilde{\Lambda}_{ijkl}^{(1)}(\vec{n})|_{\text{JL}} + \tilde{\Lambda}_{ijkl}^{(2)}(\vec{n})|_{\text{JL}}$ as follows:

$$\tilde{\Lambda}_{ijkl}(\vec{n}) = \tilde{\Lambda}_{ijkl}^{(1)}(\vec{n}) + \tilde{\Lambda}_{ijkl}^{(2)}(\vec{n}) \\ \tilde{\Lambda}_{ijkl}^{(1)}(\vec{n}) = \frac{1}{4} \{ (\delta_{jl} - 3n_j n_l) \delta_{ik} + (\delta_{jk} - 3n_j n_k) \delta_{il} \\ + (\delta_{ik} - 3n_i n_k) \delta_{jl} + (\delta_{il} - 3n_i n_l) \delta_{jk} \} \\ \tilde{\Lambda}_{ijkl}^{(2)}(\vec{n}) = \frac{1}{2} \alpha \{ -(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{jk}\delta_{il}) \\ + 3(n_i n_j \delta_{kl} + n_i n_k \delta_{jl} + n_i n_l \delta_{jk} + n_j n_k \delta_{il} + n_j n_l \delta_{ik} + n_k n_l \delta_{ij}) - 15n_i n_j n_k n_l \}$$

$$\tilde{\Lambda}_{ijkl}(\vec{n})|_{\text{JL}} = \tilde{\Lambda}_{ijkl}^{(1)}(\vec{n})|_{\text{JL}} + \tilde{\Lambda}_{ijkl}^{(2)}(\vec{n})|_{\text{JL}} \\ \tilde{\Lambda}_{ijkl}^{(1)}(\vec{n})|_{\text{JL}} = -2(\delta_{jl} - 3n_j n_l) \delta_{ik} \\ \tilde{\Lambda}_{ijkl}^{(2)}(\vec{n})|_{\text{JL}} = 2\alpha \{ -(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{jk}\delta_{il}) + 3(n_i n_j \delta_{kl} \\ + n_i n_k \delta_{jl} + n_i n_l \delta_{jk} + n_j n_k \delta_{il} + n_j n_l \delta_{ik} + n_k n_l \delta_{ij}) - 15n_i n_j n_k n_l \}. \quad (\text{A.19})$$

Here, we elucidate the differences between our result and the one derived by Joffrin and Levelut: (1) $\tilde{\Lambda}_{ijkl}^{(1)}(\vec{n})$ contains all permutation terms of the indices (jl) , (ik) and (il) , (jk) , while $\tilde{\Lambda}_{ijkl}^{(1)}(\vec{n})|_{\text{JL}}$ does not. Both $\tilde{\Lambda}_{ijkl}^{(1)}(\vec{n})$ and $\tilde{\Lambda}_{ijkl}^{(1)}(\vec{n})|_{\text{JL}}$ are correct, since the difference is originated from the definition of phonon strain field e_{ij} . According to Joffrin and Levelut, phonon strain is defined as $e_{ij} = \partial u_i / \partial x_j$, whereas our definition is $e_{ij} = \frac{1}{2}(\partial u_i / \partial x_j + \partial u_j / \partial x_i)$. Two definitions of e_{ij} differ by an anti-symmetric part $\frac{1}{2}(\partial u_i / \partial x_j - \partial u_j / \partial x_i)$. If we refer to the model of Vural and Leggett [2], this anti-symmetric part corresponds to a rotation which costs no energy. (2) Compared to our $\tilde{\Lambda}_{ijkl}^{(1)}(\vec{n})$, $\tilde{\Lambda}_{ijkl}^{(1)}(\vec{n})|_{\text{JL}}$ derived by Joffrin and Levelut missed an overall factor of $-1/2$. (3) Compared to our $\tilde{\Lambda}_{ijkl}^{(2)}(\vec{n})$, $\tilde{\Lambda}_{ijkl}^{(2)}(\vec{n})|_{\text{JL}}$ derived by them missed an overall factor of $1/4$.

Appendix B. Derivations of $\hat{H}'_2(t)$, $\hat{H}'_3(t)$, $d\mathbf{E}_\alpha^{(2)}/dt$ and $d\mathbf{E}_\alpha^{(3)}/dt$ and the proof of $d\mathbf{E}_\alpha^{(2,3)}/dt \ll d\mathbf{E}_\alpha^{(0,1)}/dt$

Now let us perform the detailed calculations of $\hat{H}'_2(t)$ and $\hat{H}'_3(t)$. The relative position $\vec{x}_s - \vec{x}_{s'}$ changes in response to external strain, leading to the change of the coefficient of many-body interaction $\Lambda_{ijkl}^{(ss')}$. Hence, the change of $\Lambda_{ijkl}^{(ss')}$ generates a time-dependent perturbation to super block Hamiltonian,

$$\hat{H}'_2(t) = \sum_{s \neq s'} \sum_{ijkl} \Delta \Lambda_{ijkl}^{(ss')} (t) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}. \quad (\text{B.1})$$

To the first order of external strain, $\Delta \Lambda_{ijkl}^{(ss')}$ is given as follows,

$$\Delta \Lambda_{ijkl}^{(ss')} = \left(\Delta \tilde{\Lambda}_{ijkl}^{(ss')} - 3\tilde{\Lambda}_{ijkl}^{(ss')} \frac{\Delta \vec{x}_{ss'} \cdot (\vec{x}_s - \vec{x}'_s)}{|\Delta \vec{x}_{ss'}| \cdot |\vec{x}_s - \vec{x}'_s|} \right) \frac{|\Delta \vec{x}_{ss'}|}{|\vec{x}_s - \vec{x}'_s|^4}, \quad (\text{B.2})$$

where $\Delta\tilde{\Lambda}_{ijkl}^{(ss')}$ is

$$\begin{aligned} \Delta\tilde{\Lambda}_{ijkl}^{(ss')} = & \left\{ \frac{3}{4} \left[2(n_j n_l \delta_{ik} + n_j n_k \delta_{il} \right. \right. \\ & + n_i n_k \delta_{jl} + n_i n_l \delta_{jk}) \frac{\Delta\vec{x}_{ss'} \cdot (\vec{x}_s - \vec{x}_s')}{|\Delta\vec{x}_{ss'}| \cdot |\vec{x}_s - \vec{x}_s'|} \\ & - [(m_j n_l + m_l n_j) \delta_{ik} + (m_j n_k + m_k n_j) \delta_{il} + (m_i n_k + m_k n_i) \delta_{jl} \\ & + (m_i n_l + m_l n_i) \delta_{jk}] \\ & - 3\alpha \left(n_k n_l \delta_{ij} + n_j n_l \delta_{ik} + n_k n_j \delta_{il} + n_i n_l \delta_{jk} + n_i n_k \delta_{jl} + n_i n_j \delta_{kl} \right) \\ & \frac{\Delta\vec{x}_{ss'} \cdot (\vec{x}_s - \vec{x}_s')}{|\Delta\vec{x}_{ss'}| \cdot |\vec{x}_s - \vec{x}_s'|} \\ & + \frac{3}{2} \alpha \left[m_i (n_l \delta_{jk} + n_k \delta_{jl} + n_j \delta_{kl}) \right. \\ & + m_j (n_l \delta_{ik} + n_k \delta_{il} + n_i \delta_{kl}) + m_k (n_l \delta_{ij} + n_i \delta_{jl} + n_j \delta_{il}) \\ & + m_l (n_k \delta_{ij} + n_i \delta_{jk} + n_j \delta_{ik}) \left. \right] \\ & - \frac{15}{2} \alpha \left(m_i n_j n_k n_l + m_j n_i n_k n_l + m_k n_i n_j n_l + m_l n_i n_j n_k \right) \\ & \left. + 30\alpha n_i n_j n_k n_l \frac{\Delta\vec{x}_{ss'} \cdot (\vec{x}_s - \vec{x}_s')}{|\Delta\vec{x}_{ss'}| \cdot |\vec{x}_s - \vec{x}_s'|} \right\}. \end{aligned} \quad (\text{B.3})$$

In equation (B.3), $\alpha = 1 - c_t^2/c_l^2$, $\Delta\vec{x}_{ss'} = \vec{u}^{(s)}(t) - \vec{u}^{(s')}(t)$, and $\vec{m} = \Delta\vec{x}_{ss'}/|\Delta\vec{x}_{ss'}|$ is the unit vector of $\Delta\vec{x}_{ss'}$.

External strain could probably alter stress tensor $\hat{T}_{ij}^{(s)}$ as well. The strain field dependence of $\hat{T}_{ij}^{(s)}$ originates from higher order expansions of the glass non-harmonic Hamiltonian (recall equation (2.12)). Since we have no idea how stress tensor changes, we qualitatively expand it to the first order of strain field: $\Delta\hat{T}_{ij}^{(s)}(t) \sim e(t)\hat{T}_{ij}^{(s)}$. This brings in another time-dependent perturbation,

$$\hat{H}'_3(t) = \sum_{s \neq s'} \sum_{ijkl} 2\Lambda_{ijkl}^{(ss')} \Delta\hat{T}_{ij}^{(s)}(t) \hat{T}_{kl}^{(s')}. \quad (\text{B.4})$$

The resonance energy absorption contribution $dE_\alpha^{(2)}(L)/dt$ comes from the second order perturbation of $\hat{H}'_2(t)$:

$$\begin{aligned} dE_l^{(2)}(L)/dt = & (1 - e^{-\beta\omega}) \frac{A^2 k^2 N_0^3 \ln N_0}{40\pi^3 (\rho c_l^2)^2} \\ & [(55 + 176\alpha + 688\alpha^2) + 44(1 + 4\alpha + 4\alpha^2)x(\omega)] \omega \\ & \int \text{Im } \tilde{\chi}_l(\Omega, L) \text{Im } \tilde{\chi}_l(\omega - \Omega, L) d\Omega \\ dE_t^{(2)}(L)/dt = & (1 - e^{-\beta\omega}) \frac{A^2 k^2 N_0^3 \ln N_0}{40\pi^3 (\rho c_t^2)^2} \\ & [(35 + 112\alpha + 656\alpha^2) + 28(1 + 4\alpha + 4\alpha^2)x(\omega)] \omega \\ & \int \text{Im } \tilde{\chi}_t(\Omega, L) \text{Im } \tilde{\chi}_t(\omega - \Omega, L) d\Omega, \end{aligned} \quad (\text{B.5})$$

where $x(\omega) = \frac{\text{Im } \tilde{\chi}_l(\omega, L)}{\text{Im } \tilde{\chi}_l(\omega, L)} - 2$. The energy absorption contribution $dE_\alpha^{(3)}(L)/dt$ comes from the second order perturbation of $\hat{H}'_3(t)$. The qualitative result for this term is

$$\begin{aligned} dE_\alpha^{(3)}(L)/dt \sim & g_\alpha (1 - e^{-\beta\omega}) \frac{A^2 k^2 N_0^3 \ln N_0}{\pi^3 (\rho c_t^2)^2} \omega \\ & \int \text{Im } \tilde{\chi}_t(\Omega, L) \text{Im } \tilde{\chi}_t(\omega - \Omega, L) d\Omega, \end{aligned} \quad (\text{B.6})$$

where $g_{\alpha=l,t}$ are dimensionless constants of order $\sim \mathcal{O}(1)$. Please note that in this paper we always expand the energy absorption terms up to the second order of non-harmonic susceptibilities. Therefore, in $dE_\alpha^{(2)}/dt$ and $dE_\alpha^{(3)}/dt$ we keep the leading order of the wave functions: $|n^{\text{sup}}\rangle \approx |n\rangle$. Furthermore, we only keep terms with $\langle \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \hat{T}_{ab}^{(u=s)} \hat{T}_{cd}^{(u'=s')} \rangle$ and $\langle \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \hat{T}_{ab}^{(u=s')} \hat{T}_{cd}^{(u'=s)} \rangle$ due to the approximation made in equation (3.2). We rewrite $dE_\alpha^{(2)}/dt$ and $dE_\alpha^{(3)}/dt$ in terms of non-harmonic susceptibilities. The coefficient $\ln N_0$ comes from the integral $\int_L^{N_0 L} d^3 r / r^3$. Together with equations (3.3) and (3.4), the total energy absorption of super block is the summation of four terms,

$$dE_\alpha^{\text{sup}}(L)/dt = dE_\alpha^{(0)}(L)/dt + dE_\alpha^{(1)}(L)/dt + dE_\alpha^{(2)}(L)/dt + dE_\alpha^{(3)}(L)/dt. \quad (\text{B.7})$$

Next, we prove that both of equations (B.5) and (B.6) are negligible compared to equations (3.3) and (3.4). To qualitatively estimate the ratio between $dE_\alpha^{(2,3)}(L)/dt$ and $dE_\alpha^{(0,1)}(L)/dt$, we assume $\text{Im } \tilde{\chi}_\alpha(\omega, L)$ with $\alpha = l, t$ are roughly independent of the frequency ω up to the upper cut-off frequency ω_c , where ω_c will be specified later.

$$\frac{dE_\alpha^{(2,3)}(L)/dt}{dE_\alpha^{(0,1)}(L)/dt} \sim \frac{1}{\rho c_\alpha^2 L^3} \frac{\omega_c}{\ln(\omega_c/\omega)}. \quad (\text{B.8})$$

From equation (B.8), there is a critical length scale L_c , below which $dE_\alpha^{(2,3)}(L)/dt$ are greater than $dE_\alpha^{(0,1)}(L)/dt$. To estimate the upper limit of L_c , we let ω_c to be an unreasonably high value 10^{15} rad/s, corresponding to an unreasonably high temperature $T_c = (\hbar\omega_c/k_B) \sim 10^4$ K. Even though we allow ω_c to take such a huge value, L_c is still much smaller than $L_1 \sim 50$ Å, the minimal length scale of elementary block suggested by Vural and Leggett [2]:

$$L_c = \left(\frac{1}{\rho c_\alpha^2} \frac{\hbar\omega_c}{\ln(\omega_c/\omega)} \right)^{\frac{1}{3}} \approx 1.7 \text{ \AA} \ll L_1 = 50 \text{ \AA}. \quad (\text{B.9})$$

The above analysis demonstrates that within the length scale of our model (i.e. $L \geq L_1 \sim 50$ Å), $dE_\alpha^{(2,3)}(L)/dt$ are always negligible compared to $dE_\alpha^{(0,1)}(L)/dt$.

Appendix C. Calculation details of resonance energy absorption $dE_\alpha^{(0)}/dt$ and $dE_\alpha^{(1)}/dt$

In this appendix, we write down the explicit form of the energy absorption rate $dE_\alpha^{(0)}/dt$ and $dE_\alpha^{(1)}/dt$ (equations (3.3) and (3.4) in the main text).

To get equation (3.1) we consider elementary block with the side length L and the non-harmonic part of Hamiltonian, \hat{H}_0 . We denote $\{|n\rangle\}$ and $\{E_n^{(s)}\}$ to be the complete and orthogonal set of eigenstates and corresponding eigenvalues of \hat{H}_0 . By adding a weak external phonon strain field $e_{ij}(t) = e_{ij} e^{-i\omega t}$,

we introduce a time-dependent perturbation $\sum_{ij} e_{ij}(t)\hat{T}_{ij}$ into the Hamiltonian: $\hat{H}_0(t) = \hat{H}_0 + \sum_{ij} e_{ij}(t)\hat{T}_{ij}$. We use the interaction picture to describe the state vectors and operators by separating $\hat{H}_0(t)$ into two parts: the time-independent part \hat{H}_0 and the time-dependent part $\sum_{ij} e_{ij}(t)\hat{T}_{ij}$. For an arbitrary operator \hat{A} and state $|\psi, t\rangle$, in the interaction picture they are defined as

$$\hat{A}_I(t) = e^{i\hat{H}_0 t} \hat{A} e^{-i\hat{H}_0 t} \quad |\psi_I, t\rangle = \mathcal{T} e^{-i \int_{-\infty}^t \sum_{ij} e_{ij}(t') \hat{T}_{ij}(t') dt'} |\psi_I, -\infty\rangle, \quad (\text{C.1})$$

where we set $\hbar = 1$. The resonance phonon energy absorption per unit time for elementary block glass is given as follows,

$$\frac{dE_\alpha}{dt} = \frac{\partial}{\partial t} \sum_n P_n \left(\langle n_I, t | \hat{H}_0 | n_I, t \rangle - \langle n_I, -\infty | \hat{H}_0 | n_I, -\infty \rangle \right), \quad (\text{C.2})$$

where $P_n = e^{-\beta E_n} / \mathcal{Z}$ is the probability function of the n th state, and $\mathcal{Z} = \sum_m e^{-\beta E_m}$ is the partition function. By assuming $\sum_{ij} e_{ij}(t)\hat{T}_{ij} \ll \hat{H}_0$, we perturbatively expand the wave vectors up to the second order of $\sum_{ij} e_{ij}(t)\hat{T}_{ij}$ to calculate the energy absorption rate for elementary block:

$$\begin{aligned} \frac{dE_\alpha}{dt} &= 2\pi\omega \sum_{nm} P_n |\langle m | \sum_{ij} e_{ij} \hat{T}_{ij} | n \rangle|^2 \delta(E_n - E_m - \omega) \\ &= 2L^3 A^2 k^2 \omega (1 - e^{-\beta\omega}) \text{Im} \tilde{\chi}_\alpha(\omega). \end{aligned} \quad (\text{C.3})$$

Equations (C.1)–(C.3) provide details on the derivation of equation (3.1). A and k are the amplitude and the wave number of external strain field, respectively. $\text{Im} \tilde{\chi}_{I,t}(\omega)$ is the ‘imaginary part of reduced non-harmonic susceptibility’ defined in equations (2.16) and (2.17). This result is equivalent to the result using Fermi’s golden rule.

We then discuss the details of computing resonance phonon energy absorption of super block, equations (3.3), (B.5), (B.6) and (3.4). We combine N_0^3 elementary blocks with the side length L to form a super block with the side length N_0L . The super block Hamiltonian $\hat{H}_0^{\text{sup}} = \hat{H}_0 + \hat{V}$, where $\hat{H}_0 = \sum_s \hat{H}_0^{(s)}$ and $\hat{V} = \sum_{s \neq s'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}$. We denote $\{n^{(s)}\}$ and $\{E_n^{(s)}\}$ to be the complete and orthogonal set of eigenstates and corresponding eigenvalues of the s th elementary block Hamiltonian $\hat{H}_0^{(s)}$. Then $|n\rangle = \prod_s |n^{(s)}\rangle$ and $E_n = \sum_s E_n^{(s)}$ are the eigenstates and eigenvalues of $\hat{H}_0 = \sum_s \hat{H}_0^{(s)}$. We also denote $|n^{\text{sup}}\rangle$ and E_n^{sup} to be the eigenstates and eigenvalues of \hat{H}_0^{sup} . By assuming $\hat{V} \ll \hat{H}_0$, the relation between $|n^{\text{sup}}\rangle$, E_n^{sup} and $|n\rangle$, E_n can be expressed as follows,

$$\begin{aligned} |n^{\text{sup}}\rangle &= |n\rangle + \sum_{p \neq n} \frac{\langle p | \hat{V} | n \rangle}{E_n - E_p} |p\rangle + \mathcal{O}(V^2) \\ E_n^{\text{sup}} &= E_n + \langle n | \hat{V} | n \rangle + \mathcal{O}(V^2). \end{aligned} \quad (\text{C.4})$$

Here, we put in a weak external strain field $e_{ij}^{(s)}(t) = e_{ij}^{(s)} e^{-i\omega t}$. As we have discussed in section 2.2 in the main text, super block Hamiltonian receives a time-dependent perturbation $\hat{H}'(t)$. Finally, the total Hamiltonian of super block is summarized as follows,

$$\begin{aligned} \hat{H}_0^{\text{sup}}(t) &= \hat{H}_0^{\text{sup}} + \hat{H}'(t) \\ \hat{H}'(t) &= \sum_{s=1}^{N_0^3} \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)} \\ &+ \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \left(\Delta \Lambda_{ijkl}^{(ss')} (t) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} + 2\Lambda_{ijkl}^{(ss')} \Delta \hat{T}_{ij}^{(s)}(t) \hat{T}_{kl}^{(s')} \right). \end{aligned} \quad (\text{C.5})$$

Let us now use the interaction picture to describe the state vectors and operators by dividing $\hat{H}_0^{\text{sup}}(t)$ into two parts, the time-independent part \hat{H}_0^{sup} and the time-dependent part $\hat{H}'(t)$. For an arbitrary operator \hat{B} and state $|\phi, t\rangle$, in the interaction picture they are defined as

$$\hat{B}_I(t) = e^{i\hat{H}_0^{\text{sup}} t} \hat{B} e^{-i\hat{H}_0^{\text{sup}} t} \quad |\phi_I, t\rangle = \mathcal{T} e^{-i \int_{-\infty}^t H'(t') dt'} |\phi_I, -\infty\rangle. \quad (\text{C.6})$$

The resonance phonon energy absorption per unit time for super block glass is given by

$$\frac{dE_\alpha^{\text{sup}}}{dt} = \frac{\partial}{\partial t} \sum_n P_n^{\text{sup}} \left(\langle n_I^{\text{sup}}, t | \hat{H}_0^{\text{sup}} | n_I^{\text{sup}}, t \rangle - \langle n_I^{\text{sup}}, -\infty | \hat{H}_0^{\text{sup}} | n_I^{\text{sup}}, -\infty \rangle \right), \quad (\text{C.7})$$

where $P_n^{\text{sup}} = e^{-\beta E_n^{\text{sup}}} / \mathcal{Z}^{\text{sup}}$ is the probability function. Similarly, we expand the wave functions in equation (C.7) up to the second order of $\hat{H}'(t)$ to calculate resonance phonon energy absorption. Among various terms in the expansions, we drop the following terms: (1) terms with odd numbers of strain fields $e_{ij}(t)$, because one period of time average makes such terms to vanish; (2) terms with odd numbers of non-harmonic stress tensors, because these terms contain odd numbers of the diagonal matrix element of non-harmonic stress tensor operator, $\langle n | \hat{T}_{ij}^{(s)} | n \rangle \propto \delta \langle \hat{H}^{(s)} \rangle / \delta e_{ij}$, which is the ‘total’ stress tensor minus the elastic stress tensor. It is highly plausible that the expectation value of the non-harmonic stress tensor tends to vanish for large enough block of glass. Based on these arguments, we keep the non-vanishing terms as follows,

$$\begin{aligned} \frac{dE_\alpha^{\text{sup}}}{dt} &= \frac{\partial}{\partial t} \sum_n P_n^{\text{sup}} \mathcal{T} \int_{-\infty}^t dt' dt'' \sum_{j^{\text{sup}}} e^{-i(E_{I'}^{\text{sup}} - E_n^{\text{sup}})(t' - t'')} (E_{I'}^{\text{sup}} - E_n^{\text{sup}}) \\ &\left\{ \sum_{ss'} \sum_{ijkl} \langle n^{\text{sup}} | e_{ij}^{(s)}(t') \hat{T}_{ij}^{(s)} | I^{\text{sup}} \rangle \langle I^{\text{sup}} | e_{kl}^{(s')}(t'') \hat{T}_{kl}^{(s')} | n^{\text{sup}} \rangle \right. \\ &+ \sum_{s \neq s'} \sum_{u \neq u'} \sum_{abcdijkl} \langle n^{\text{sup}} | \Delta \Lambda_{ijkl}^{(ss')} (t) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} | I^{\text{sup}} \rangle \langle I^{\text{sup}} | \Delta \Lambda_{abcd}^{(uu')} (t) \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | n^{\text{sup}} \rangle \\ &\left. + \sum_{s \neq s'} \sum_{u \neq u'} \sum_{abcdijkl} \langle n^{\text{sup}} | 2\Lambda_{ijkl}^{(ss')} \Delta \hat{T}_{ij}^{(s)}(t) \hat{T}_{kl}^{(s')} | I^{\text{sup}} \rangle \langle I^{\text{sup}} | 2\Lambda_{abcd}^{(uu')} \Delta \hat{T}_{ab}^{(u)}(t) \hat{T}_{cd}^{(u')} | n^{\text{sup}} \rangle \right\}, \end{aligned} \quad (\text{C.8})$$

where $dE_\alpha^{(2)}/dt$ and $dE_\alpha^{(3)}/dt$ have been already derived in appendix B. Now let us focus on the first term on the right-hand side of the above equation, which we denote as letter J . The purpose of this appendix is to calculate J , which we will later show that it is the summation of $dE_\alpha^{(0)}/dt$ and $dE_\alpha^{(1)}/dt$. We integrate over the time variables t', t'' to simplify J as follows:

$$J = 2\pi\omega (1 - e^{-\beta\omega}) \sum_{n^{\text{sup}}} P_n^{\text{sup}} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \sum_{l^{\text{sup}}} \langle n^{\text{sup}} | \hat{T}_{ij}^{(s)} | l^{\text{sup}} \rangle \langle l^{\text{sup}} | \hat{T}_{kl}^{(s')} | n^{\text{sup}} \rangle \delta(\omega - E_l^{\text{sup}} + E_n^{\text{sup}}), \quad (\text{C.9})$$

where $e_{ij} = \frac{1}{2}(A_i k_j + A_j k_i)$. To expand J up to the second order of the susceptibilities, we expand the wave functions and the energy eigenvalues to the first order of \hat{V} : $|n^{\text{sup}}\rangle \approx |n\rangle + \sum_{p \neq n} \frac{\langle p | \hat{V} | n \rangle}{E_n - E_p} |p\rangle$, $E_n^{\text{sup}} = E_n + \langle n | \hat{V} | n \rangle$:

$$\begin{aligned} J &= 2\pi\omega (1 - e^{-\beta\omega}) \sum_{nl} P_n \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \delta(\omega - E_l + E_n) \langle n | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | n \rangle & \frac{dE_\alpha^{(0)}}{dt} \\ &- 2\pi\omega (1 - e^{-\beta\omega}) \sum_{nl} P_n \beta \langle n | \hat{V} | n \rangle \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \delta(\omega - E_l + E_n) \langle n | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | n \rangle & J_1 \\ &+ 2\pi\omega (1 - e^{-\beta\omega}) \sum_{nml} P_n P_m \beta \langle m | \hat{V} | m \rangle \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \delta(\omega - E_l + E_n) \langle n | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | n \rangle & J_2 \\ &+ 2\pi\omega (1 - e^{-\beta\omega}) \sum_{nml} P_n \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \delta(\omega - E_l + E_n) \frac{\langle n | \hat{V} | m \rangle}{E_n - E_m} \langle m | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | n \rangle & J_3 \\ &+ 2\pi\omega (1 - e^{-\beta\omega}) \sum_{nlp} P_n \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \delta(\omega - E_l + E_n) \langle n | \hat{T}_{ij}^{(s)} | p \rangle \frac{\langle p | \hat{V} | l \rangle}{E_l - E_p} \langle l | \hat{T}_{kl}^{(s')} | n \rangle & J_4 \\ &+ 2\pi\omega (1 - e^{-\beta\omega}) \sum_{nlN} P_n \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \delta(\omega - E_l + E_n) \langle n | \hat{T}_{ij}^{(s)} | l \rangle \frac{\langle l | \hat{V} | N \rangle}{E_l - E_N} \langle N | \hat{T}_{kl}^{(s')} | n \rangle & J_5 \\ &+ 2\pi\omega (1 - e^{-\beta\omega}) \sum_{nlM} P_n \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \delta(\omega - E_l + E_n) \langle n | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | M \rangle \frac{\langle M | \hat{V} | n \rangle}{E_n - E_M}. & J_6 \quad (\text{C.10}) \end{aligned}$$

By using the approximation in equation (3.2), we prove that $dE_\alpha^{(0)}/dt$ is simply the summation of elementary block resonance energy absorption (equation (3.3)):

$$\frac{dE_\alpha^{(0)}}{dt} = 2N_0^3 L^3 A^2 k^2 \omega (1 - e^{-\beta\omega}) \text{Im} \tilde{\chi}_\alpha(\omega). \quad (\text{C.11})$$

Meanwhile, the summation of J_1, J_2, J_3, J_4, J_5 and J_6 lead to $dE_\alpha^{(1)}/dt$ (equation (3.4)). With the exchange of indices, we have $J_5 = J_3$ and $J_6 = J_4$. Therefore, only J_1, J_2, J_3 and J_4 need to be calculated, which we explicitly show the steps in the following part.

To calculate J_1 , we replace $\langle n | \hat{V} | n \rangle$ with $\sum_{abcd} \sum_{u \neq u'} \Lambda_{abcd}^{(uu')} \langle n | \hat{T}_{ab}^{(u)} | m \rangle \langle m | \hat{T}_{cd}^{(u')} | n \rangle$ and obtain J_1 as follows,

$$J_1 = -2\pi\omega (1 - e^{-\beta\omega}) \sum_{nml} \sum_{abcdijkl} \sum_{u \neq u', ss'} P_n \beta \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \delta(\omega - E_l + E_n) \langle n | \hat{T}_{ab}^{(u)} | m \rangle \langle m | \hat{T}_{cd}^{(u')} | n \rangle \langle n | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | n \rangle. \quad (\text{C.12})$$

From equation (3.2), the product of the stress tensors in different block space, $\langle n | \hat{T}_{ab}^{(u)} | l \rangle \langle l | \hat{T}_{cd}^{(u')} | n \rangle_{u \neq u'} \approx 0$, because the stress tensors in different block space (i.e. $\mathbf{T} = \delta \hat{H}^{(s)} / \delta \mathbf{e}$) are random and uncorrelated. After averaging over the entire block space, terms like $\langle n | \hat{T}_{ab}(\vec{x}_u) | l \rangle \langle l | \hat{T}_{cd}(\vec{x}'_u) | n \rangle_{u \neq u'}$ will cancel with each other. Therefore, we only keep the product of stress tensors belonging to the same block space, which means only the terms like $\langle n | \hat{T}_{ab}^{(u)} | l \rangle \langle l | \hat{T}_{cd}^{(u')} | n \rangle_{u=u'}$ do not vanish. Based on the above argument, the upper indices of the stress tensors in equation (C.12) have to be 'labelled in pairs'. To be more specific, we set $s = u$ and $s' = u'$, the stress tensors in equation (C.12) are written as:

$\langle n | \hat{T}_{ab}^{(u)} | m \rangle \langle m | \hat{T}_{cd}^{(u')} | n \rangle \langle n | \hat{T}_{ij}^{(s=u)} | l \rangle \langle l | \hat{T}_{kl}^{(s'=u')} | n \rangle$. Another non-zero case is that we set $s = u'$ and $s' = u$. Hereby, J_1 can be simplified into two terms:

$$J_1 = -2\pi\omega (1 - e^{-\beta\omega}) \sum_{nml} \sum_{abcdijkl} P_n \beta e_{ij} e_{kl} \delta(\omega - E_l + E_n) \langle n | \hat{T}_{ab}^{(u)} | m \rangle \langle m | \hat{T}_{cd}^{(u')} | n \rangle \sum_{u \neq u'} e^{-i\vec{k}\cdot(\vec{x}_u - \vec{x}'_u)} \Lambda_{abcd}^{(uu')} \left(\langle n | \hat{T}_{ij}^{(u)} | l \rangle \langle l | \hat{T}_{kl}^{(u')} | n \rangle + \langle n | \hat{T}_{ij}^{(u')} | l \rangle \langle l | \hat{T}_{kl}^{(u)} | n \rangle \right). \quad (\text{C.13})$$

Upon the symmetry property $\Lambda_{abcd}^{(uu')} = \Lambda_{abcd}^{(u'u)}$, the second term in equation (C.13) equals to the first term. We continue calculating J_1 as follows,

$$\begin{aligned}
 J_1 &= -4\pi\omega (1 - e^{-\beta\omega}) \sum_{abcdijkl} \sum_{u \neq u'} \sum_{n^{(u)} n^{(u')} m^{(u)} m^{(u')} l^{(u)} l^{(u')}} P_n \beta \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \delta(\omega - E_l + E_n) \\
 &\quad \langle n^{(u)} | \hat{T}_{ab}^{(u)} | m^{(u)} \rangle \langle m^{(u)} | n^{(u)} \rangle \langle n^{(u)} | \hat{T}_{ij}^{(u)} | l^{(u)} \rangle \langle l^{(u)} | n^{(u)} \rangle \langle n^{(u')} | m^{(u')} \rangle \langle m^{(u')} | \hat{T}_{cd}^{(u')} | n^{(u')} \rangle \langle n^{(u')} | l^{(u')} \rangle \langle l^{(u')} | \hat{T}_{kl}^{(u')} | n^{(u')} \rangle \\
 &= -4\pi\omega (1 - e^{-\beta\omega}) \sum_{abcdijkl} \sum_{u \neq u'} \sum_{n^{(u)} n^{(u')} m^{(u)} m^{(u')} l^{(u)} l^{(u')}} P_n \beta \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \delta(\omega - E_l + E_n) \\
 &\quad \langle n^{(u)} | \hat{T}_{ab}^{(u)} | m^{(u)} \rangle \langle n^{(u)} | \hat{T}_{ij}^{(u)} | l^{(u)} \rangle \langle m^{(u')} | \hat{T}_{cd}^{(u')} | n^{(u')} \rangle \langle l^{(u')} | \hat{T}_{kl}^{(u')} | n^{(u')} \rangle \delta_{ln} \delta_{lm} \\
 &= 0.
 \end{aligned} \tag{C.14}$$

In the first step of equation (C.14), we use the definition $|n\rangle = \prod_s |n^{(s)}\rangle$ to decompose the wave functions. In the second step, because of $n = l$, we have $E_n = E_l$ which renders the δ -function to vanish.

Similar approach is applied to calculate J_2 : we replace $\langle m | \hat{V} | m \rangle$ with $\sum_{abcd} \sum_{u \neq u'} \sum_p \Lambda_{abcd}^{(uu')} \langle n | \hat{T}_{ab}^{(u)} | p \rangle \langle p | \hat{T}_{cd}^{(u')} | n \rangle$, and use the same steps in calculating J_1 :

$$\begin{aligned}
 J_2 &= 2\pi\omega (1 - e^{-\beta\omega}) \sum_{nmlp} \sum_{abcdijkl} \sum_{u \neq u', ss'} P_n P_m \beta \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \delta(\omega - E_l + E_n) \\
 &\quad \langle m | \hat{T}_{ab}^{(u)} | p \rangle \langle p | \hat{T}_{cd}^{(u')} | m \rangle \langle n | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | n \rangle \\
 &= 4\pi\omega (1 - e^{-\beta\omega}) \sum_{nmlp} \sum_{abcdijkl} \sum_{u \neq u'} P_n P_m \beta \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \delta(\omega - E_l + E_n) \\
 &\quad \langle m | \hat{T}_{ab}^{(u)} | p \rangle \langle p | \hat{T}_{cd}^{(u')} | m \rangle \langle n | \hat{T}_{ij}^{(u)} | l \rangle \langle l | \hat{T}_{kl}^{(u')} | n \rangle \\
 &= 4\pi\omega (1 - e^{-\beta\omega}) \sum_{nmlp} \sum_{abcdijkl} \sum_{u \neq u'} P_n P_m \beta \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \delta(\omega - E_l + E_n) \\
 &\quad \langle m^{(u)} | \hat{T}_{ab}^{(u)} | p^{(u)} \rangle \langle p^{(u)} | m^{(u)} \rangle \langle n^{(u)} | \hat{T}_{ij}^{(u)} | l^{(u)} \rangle \langle l^{(u)} | n^{(u)} \rangle \langle m^{(u')} | p^{(u')} \rangle \langle p^{(u')} | \hat{T}_{cd}^{(u')} | m^{(u')} \rangle \langle n^{(u')} | l^{(u')} \rangle \langle l^{(u')} | \hat{T}_{kl}^{(u')} | n^{(u')} \rangle \\
 &= 4\pi\omega (1 - e^{-\beta\omega}) \sum_{nmlp} \sum_{abcdijkl} \sum_{u \neq u'} P_n P_m \beta \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \delta(\omega - E_l + E_n) \\
 &\quad \langle m^{(u)} | \hat{T}_{ab}^{(u)} | p^{(u)} \rangle \langle n^{(u)} | \hat{T}_{ij}^{(u)} | l^{(u)} \rangle \langle p^{(u')} | \hat{T}_{cd}^{(u')} | m^{(u')} \rangle \langle l^{(u')} | \hat{T}_{kl}^{(u')} | n^{(u')} \rangle \delta_{lm} \delta_{pm} \\
 &= 0.
 \end{aligned} \tag{C.15}$$

Next, we perform the derivation of J_3 : we replace $\langle n | \hat{V} | m \rangle$ with $\sum_{abcd} \sum_{u \neq u'} \sum_p \Lambda_{abcd}^{(uu')} \langle n | \hat{T}_{ab}^{(u)} | p \rangle \langle p | \hat{T}_{cd}^{(u')} | m \rangle$ to express J_3 as follows,

$$\begin{aligned}
 J_3 &= 4\pi\omega (1 - e^{-\beta\omega}) \sum_n P_n \sum_{ijklabcd} \sum_{u \neq u'} \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \\
 &\quad \sum_{lmp} \frac{\delta(\omega - E_l + E_n)}{E_n - E_m} \langle n | \hat{T}_{ab}^{(u)} | p \rangle \langle p | \hat{T}_{cd}^{(u')} | m \rangle \langle m | \hat{T}_{ij}^{(u)} | l \rangle \langle l | \hat{T}_{kl}^{(u')} | n \rangle \\
 &= 4\pi\omega (1 - e^{-\beta\omega}) \sum_n P_n \sum_{ijklabcd} \sum_{u \neq u'} \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \sum_{\substack{l^{(u)} l^{(u')} m^{(u)} \\ m^{(u')} p^{(u)} p^{(u')}} \frac{\delta(\omega - E_l^{(u)} - E_l^{(u')} + E_n^{(u)} + E_n^{(u')})}{E_n^{(u)} + E_n^{(u')} - E_m^{(u)} - E_m^{(u')}} \\
 &\quad \langle n^{(u)} | \hat{T}_{ab}^{(u)} | p^{(u)} \rangle \langle p^{(u)} | m^{(u)} \rangle \langle m^{(u)} | \hat{T}_{ij}^{(u)} | l^{(u)} \rangle \langle l^{(u)} | n^{(u)} \rangle \langle n^{(u')} | p^{(u')} \rangle \langle p^{(u')} | \hat{T}_{cd}^{(u')} | m^{(u')} \rangle \langle m^{(u')} | l^{(u')} \rangle \langle l^{(u')} | \hat{T}_{kl}^{(u')} | n^{(u')} \rangle.
 \end{aligned} \tag{C.16}$$

In the first step of equation (C.16), we set $s = u, s' = u'$ or $s = u', s' = u$ to label the upper indices of the stress tensors in pairs. Again we use the symmetry property $\Lambda_{abcd}^{(uu')} = \Lambda_{abcd}^{(u'u)}$ to simplify the result. In the second step of equation (C.16), we use the identity $|n\rangle = \prod_s |n^{(s)}\rangle$ and $E_n = \sum_s E_n^{(s)}$ to decompose the wave function and energy eigenvalues. We arrive at

$$\begin{aligned}
J_3 &= 4\pi\omega (1 - e^{-\beta\omega}) \sum_{n^{(u)}n^{(u')}} P_{n^{(u)}} P_{n^{(u')}} \sum_{ijklabcd} \sum_{u \neq u'} \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \\
&\quad \sum_{m^{(u)}m^{(u')}} \frac{\delta(\omega - E_m^{(u')} + E_n^{(u')})}{E_n^{(u)} + E_n^{(u')} - E_m^{(u)} - E_m^{(u')}} \langle n^{(u)} | \hat{T}_{ab}^{(u)} | m^{(u)} \rangle \langle m^{(u)} | \hat{T}_{ij}^{(u)} | n^{(u)} \rangle \langle n^{(u')} | \hat{T}_{cd}^{(u')} | m^{(u')} \rangle \langle m^{(u')} | \hat{T}_{kl}^{(u')} | n^{(u')} \rangle \\
&= 4\pi\omega (1 - e^{-\beta\omega}) \sum_{n^{(u)}n^{(u')}} P_{n^{(u)}} P_{n^{(u')}} \sum_{ijklabcd} \sum_{u \neq u'} \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \\
&\quad \sum_{m^{(u)}m^{(u')}} \frac{\delta(\omega - E_m^{(u')} + E_n^{(u')})}{E_n^{(u)} - E_m^{(u)} - \omega} \langle n^{(u)} | \hat{T}_{ab}^{(u)} | m^{(u)} \rangle \langle m^{(u)} | \hat{T}_{ij}^{(u)} | n^{(u)} \rangle \langle n^{(u')} | \hat{T}_{cd}^{(u')} | m^{(u')} \rangle \langle m^{(u')} | \hat{T}_{kl}^{(u')} | n^{(u')} \rangle \\
&= -4\omega L^3 (1 - e^{-\beta\omega}) \sum_{ijklabcd} \sum_{u \neq u'} \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \\
&\quad \sum_{n^{(u')}} P_{n^{(u')}} \text{Im} \tilde{\chi}_{cdkl}^{n^{(u')}}(\omega) \sum_{n^{(u)}m^{(u)}} P_{n^{(u)}} \frac{\langle n^{(u)} | \hat{T}_{ab}^{(u)} | m^{(u)} \rangle \langle m^{(u)} | \hat{T}_{ij}^{(u)} | n^{(u)} \rangle}{\omega - E_n^{(u)} + E_m^{(u)}} \\
&= -\frac{4\omega L^6}{\pi} (1 - e^{-\beta\omega}) \sum_{ijklabcd} \sum_{u \neq u'} \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \text{Im} \tilde{\chi}_{cdkl}(\omega) \mathcal{P} \int_0^\infty \frac{\text{Im} \tilde{\chi}_{ijab}(\Omega) d\Omega}{\omega + \Omega}. \tag{C.17}
\end{aligned}$$

In the first step of equation (C.17), we make use of the Kronecker δ -functions to reduce the summations. In the second step, we replace $E_m^{(u')} - E_n^{(u')}$ with ω in the denominator. In the third and fourth steps, we use equation (2.16) to rewrite the product of the stress tensors in terms of the susceptibility.

To obtain J_4 we replace $\langle p | \hat{V} | l \rangle$ with $\sum_{abcd} \sum_{u \neq u'} \sum_m \Lambda_{abcd}^{(uu')} \langle p | \hat{T}_{ab}^{(u)} | m \rangle \langle m | \hat{T}_{cd}^{(u')} | l \rangle$,

$$\begin{aligned}
J_4 &= 4\pi\omega (1 - e^{-\beta\omega}) \sum_n \sum_{abcdijkl} \sum_{u \neq u'} P_n \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \\
&\quad \sum_{lpm} \frac{\delta(\omega - E_l + E_n)}{E_l - E_p} \langle n | \hat{T}_{ij}^{(u)} | p \rangle \langle p | \hat{T}_{ab}^{(u)} | m \rangle \langle m | \hat{T}_{cd}^{(u')} | l \rangle \langle l | \hat{T}_{kl}^{(u')} | n \rangle \\
&= 4\pi\omega (1 - e^{-\beta\omega}) \sum_n \sum_{abcdijkl} \sum_{u \neq u'} P_n \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \sum_{\substack{l^{(u)}l^{(u')}p^{(u)} \\ p^{(u')}m^{(u)}m^{(u')}} \frac{\delta(\omega - E_l^{(u)} - E_l^{(u')} + E_n^{(u)} + E_n^{(u')})}{E_l^{(u)} + E_l^{(u')} - E_p^{(u)} - E_p^{(u')}} \\
&\quad \langle n^{(u)} | \hat{T}_{ij}^{(u)} | p^{(u)} \rangle \langle p^{(u)} | \hat{T}_{ab}^{(u)} | m^{(u)} \rangle \langle m^{(u)} | l^{(u)} \rangle \langle l^{(u)} | n^{(u)} \rangle \langle n^{(u')} | p^{(u')} \rangle \langle p^{(u')} | m^{(u')} \rangle \langle m^{(u')} | \hat{T}_{cd}^{(u')} | l^{(u')} \rangle \langle l^{(u')} | \hat{T}_{kl}^{(u')} | n^{(u')} \rangle \\
&= 4\pi\omega (1 - e^{-\beta\omega}) \sum_{n^{(u)}n^{(u')}} \sum_{abcdijkl} \sum_{u \neq u'} P_{n^{(u)}} P_{n^{(u')}} \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \\
&\quad \sum_{l^{(u')}p^{(u)}} \frac{\delta(\omega - E_l^{(u')} + E_n^{(u')})}{E_n^{(u)} + E_l^{(u')} - E_p^{(u)} - E_n^{(u')}} \langle n^{(u)} | \hat{T}_{ij}^{(u)} | p^{(u)} \rangle \langle p^{(u)} | \hat{T}_{ab}^{(u)} | n^{(u)} \rangle \langle n^{(u')} | \hat{T}_{cd}^{(u')} | l^{(u')} \rangle \langle l^{(u')} | \hat{T}_{kl}^{(u')} | n^{(u')} \rangle \\
&= 4\pi\omega (1 - e^{-\beta\omega}) \sum_{abcdijkl} \sum_{u \neq u'} \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \sum_{n^{(u)}p^{(u)}} P_{n^{(u)}} \frac{\langle n^{(u)} | \hat{T}_{ij}^{(u)} | p^{(u)} \rangle \langle p^{(u)} | \hat{T}_{ab}^{(u)} | n^{(u)} \rangle}{E_n^{(u)} - E_p^{(u)} + \omega} \\
&\quad \sum_{l^{(u')}n^{(u')}} P_{n^{(u')}} \delta(\omega - E_l^{(u')} + E_n^{(u')}) \langle n^{(u')} | \hat{T}_{cd}^{(u')} | l^{(u')} \rangle \langle l^{(u')} | \hat{T}_{kl}^{(u')} | n^{(u')} \rangle \\
&= -\frac{4\omega L^6}{\pi} (1 - e^{-\beta\omega}) \sum_{abcdijkl} \sum_{u \neq u'} \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \text{Im} \tilde{\chi}_{cdkl}(\omega) \mathcal{P} \int_0^\infty \frac{\text{Im} \tilde{\chi}_{ijab}(\Omega) d\Omega}{\Omega - \omega}. \tag{C.18}
\end{aligned}$$

Likewise, in the first step we set $s = u$, $s' = u'$ or $s = u'$, $s' = u$ to label the upper indices in pairs, and use the symmetry property of $\Lambda_{abcd}^{(uu')}$ to simplify the result. In the second step we use $|n\rangle = \prod_s |n^{(s)}\rangle$ and $E_n = \sum_s E_n^{(s)}$ to decompose the wave function and energy eigenvalues. In the third step we use the Kronecker δ -functions to reduce the summations. In the fourth step we recombine the stress tensors and make use of equation (2.16) to rewrite the product of the stress tensors in terms of the susceptibility. We sum equations (C.17) and (C.18) together,

$$4\omega L^6 (1 - e^{-\beta\omega}) \sum_{ijklabcd} \sum_{u \neq u'} \left[-\Lambda_{abcd}^{(uu')} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \right] e_{ij} e_{kl} \text{Im} \tilde{\chi}_{cdkl}(\omega) \left(\frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\Omega \text{Im} \tilde{\chi}_{ijab}(\Omega) d\Omega}{\Omega^2 - \omega^2} \right). \quad (\text{C.19})$$

Now we calculate the coefficient which appears in equation (C.19): $\sum_{u \neq u'} [-\Lambda_{abcd}^{(uu')} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)}]$. First we simplify it as follows,

$$\sum_{u \neq u'} \left[-\Lambda_{abcd}^{(uu')} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \right] = \frac{1}{8} \sum_{\vec{x}_u + \vec{x}'_u} \sum_{\vec{x}_u - \vec{x}'_u} \left[-\Lambda_{abcd}^{(uu')} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \right] = N_0^3 \sum_{\vec{x}_{uu'}} \left[-\Lambda_{abcd}^{(uu')} e^{-i\vec{k} \cdot \vec{x}_{uu'}} \right], \quad (\text{C.20})$$

where we denote $\vec{x}_u - \vec{x}'_u = \vec{x}_{uu'}$. Then we substitute in equation (A.12), the original form of $\Lambda_{abcd}^{(uu')}$, into equation (C.20) to further calculate it as follows,

$$\sum_{u \neq u'} \left[-\Lambda_{abcd}^{(uu')} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \right] = \frac{N_0^3}{2\rho c_t^2} \sum_{\vec{q}} \left[-\alpha \left(\frac{q_a q_b q_c q_d}{q^4} \right) + \frac{1}{4} \left(\frac{q_b q_d \delta_{ac} + q_b q_c \delta_{ad} + q_a q_d \delta_{bc} + q_a q_c \delta_{bd}}{q^2} \right) \right] \sum_{\vec{x}_{uu'}} e^{i(\vec{q} - \vec{k}) \cdot \vec{x}_{uu'}}. \quad (\text{C.21})$$

In equation (C.21) we need to be very cautious when evaluating $\sum_{\vec{x}_{uu'}} e^{i(\vec{q} - \vec{k}) \cdot \vec{x}_{uu'}}$: notice that $\vec{x}_{uu'} \neq 0$, and $N_0 L \geq |\vec{x}_{uu'}| \geq L$. Then we have

$$\sum_{\vec{x}_{uu'}, L \leq |\vec{x}_{uu'}| \leq N_0 L} e^{i(\vec{q} - \vec{k}) \cdot \vec{x}_{uu'}} = \frac{4\pi}{3} (N_0^3 - 1) \delta_{q_1, k_1} \delta_{q_2, k_2} \delta_{q_3, k_3} = \frac{4\pi}{3} [(1 + N_0 - 1)^3 - 1] \delta_{q_1, k_1} \delta_{q_2, k_2} \delta_{q_3, k_3} \approx \frac{4\pi}{3} [(1 + \ln N_0)^3 - 1] \delta_{q_1, k_1} \delta_{q_2, k_2} \delta_{q_3, k_3} \approx 4\pi \ln N_0 \delta_{q_1, k_1} \delta_{q_2, k_2} \delta_{q_3, k_3} \quad (\text{C.22})$$

where in the above calculations, we use the approximation $N_0 = 1 + (N_0 - 1) \approx 1 + \ln N_0$, and keep the first order of $\ln N_0$ to obtain the final result of equation (C.22). We plug this result back to equation (C.21), to simplify $\sum_{u \neq u'} [-\Lambda_{abcd}^{(uu')} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)}]$ as follows,

$$\sum_{u \neq u'} \left[-\Lambda_{abcd}^{(uu')} e^{-i\vec{k} \cdot (\vec{x}_u - \vec{x}'_u)} \right] = \frac{2\pi N_0^3 \ln N_0}{\rho c_t^2} \left[-\alpha \left(\frac{k_a k_b k_c k_d}{k^4} \right) + \frac{1}{4} \left(\frac{k_b k_d \delta_{ac} + k_b k_c \delta_{ad} + k_a k_d \delta_{bc} + k_a k_c \delta_{bd}}{k^2} \right) \right]. \quad (\text{C.23})$$

Finally, we plug equations (C.23) into (C.19), and use $e_{ij} = \frac{1}{2}(A_i k_j + A_j k_i)$ to sum over the indices i, j, k, l, a, b, c, d . This will give us

$$(1 - e^{-\beta\omega}) \frac{2N_0^3 L^3 A^2 k^2 \ln N_0}{\rho c_\alpha^2} \omega \text{Im} \tilde{\chi}_\alpha(\omega) \left(\frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\Omega \text{Im} \tilde{\chi}_\alpha(\Omega) d\Omega}{\Omega^2 - \omega^2} \right). \quad (\text{C.24})$$

The overall sign of equation (C.24) is positive, basically because in equation (C.23), the momentum average of $[-\Lambda_{abcd}(\vec{k})]$ is positive. A more direct explanation can be found in equation (A.8), where we derive the virtual phonon exchange interaction (together with $\Lambda_{ijkl}^{(ss')}$) by adding the extra term $'-m\omega_{\vec{q}\mu}^2 |u_\mu^{(0)}(\vec{q})|^2/2'$ after completing the square of the displacement operator. The negative sign in this extra term $'-m\omega_{\vec{q}\mu}^2 |u_\mu^{(0)}(\vec{q})|^2/2'$ basically determines the negativity of $\Lambda_{ijkl}^{(ss')}$ (the reader can refer to equations (A.10), (A.11) and (A.18) for further discussions). Therefore, $\Lambda_{abcd}(\vec{k})$ is on average negative, and the overall signs of equations (C.19) and (C.24) are positive.

Finally we add J_1 through J_6 to finally get equation (3.4):

$$\frac{dE_\alpha^{(1)}}{dt} = \sum_{n=1}^6 J_n = (1 - e^{-\beta\omega}) \frac{4N_0^3 L^3 A^2 k^2 \ln N_0}{\rho c_\alpha^2} \omega \text{Im} \tilde{\chi}_\alpha(\omega) \left(\frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\Omega \text{Im} \tilde{\chi}_\alpha(\Omega) d\Omega}{\Omega^2 - \omega^2} \right) = (1 - e^{-\beta\omega}) \frac{4N_0^3 L^3 A^2 k^2 \ln N_0}{\rho c_\alpha^2} \omega \text{Im} \tilde{\chi}_\alpha(\omega) \text{Re} \tilde{\chi}_\alpha(\omega), \quad (\text{C.25})$$

where $\text{Re} \tilde{\chi}_\alpha(\omega) = \frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\Omega \text{Im} \tilde{\chi}_\alpha(\Omega) d\Omega}{\Omega^2 - \omega^2}$ is the 'real part of the reduced non-harmonic susceptibility' obtained by the Kramers–Kronig relation.

Appendix D. Phonon energy absorption from electric dipole-dipole interaction

Besides the previous discussions, we should also consider how electric dipole-dipole interaction \hat{V}^{dipole} affects phonon energy absorption in dielectric amorphous materials. On the one hand, external phonon wave (not electromagnetic wave) can modify electric dipole moments by changing relative positions of positive-negative charges. On the other hand, external phonon wave can change the relative positions between electric dipoles at different positions. As a result, external phonons change electric dipole-dipole interaction and provide an effective time-dependent perturbation.

Later discussions show that the contribution of dipole-dipole interaction is negligible compared to equation (3.4). As we have shown in equations (B.5) and (B.6), the absorption due to the changes in stress tensor ($\Delta \hat{T}_{ij}$) and the changes in the many-body interaction coefficient ($\Delta \Lambda_{ijkl}^{(ss')}$) are renormalization irrelevant. Then we will demonstrate that the same is true for the ultrasound absorption contribution from the change of dipole moments and inter-dipolar positions.

Here we consider an elementary block of dielectric material with the Hamiltonian \hat{H}_0 , the eigenbasis $\{|n\rangle\}$ and the eigenvalues $\{E_n\}$. Similar as the definition of non-harmonic susceptibility in equation (2.14), the dielectric susceptibility is defined as follows

$$\begin{aligned} \text{Im } \chi_{ij}(\omega) &\stackrel{\text{def}}{=} (1 - e^{-\beta\omega}) \text{Im } \tilde{\chi}_{ij}(\omega) \\ \text{Im } \tilde{\chi}_{ij}(\omega) &= \frac{\pi}{L^3} \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \sum_n \langle m | \hat{p}_i | n \rangle \langle n | \hat{p}_j | m \rangle \delta(E_n - E_m - \omega). \end{aligned} \quad (\text{D.1})$$

We assume the dielectric material is isotropic and is invariant under SO(3) rotational group. Thus the dielectric susceptibility χ_{ij} yields the isotropic form $\chi_{ij} = \chi \delta_{ij}$.

To perform the renormalization process, we combine N_0^3 elementary blocks with the side length L to form a super block with the side length N_0L . We use the approximation to replace $\vec{x} - \vec{x}'$ by $\vec{x}_s - \vec{x}_{s'}$ for the pair of the s th and s' th blocks where \vec{x}_s denotes the center of the s th block, and $\int_{V(s)} \hat{p}_i(\vec{x}) d^3x = \hat{p}_i^{(s)}$ is the uniform electric dipole moment. The super block Hamiltonian of dielectric amorphous solid is given by

$$\begin{aligned} \hat{H}_0^{\text{sup}} &= \hat{H}_0 + \hat{V} + \hat{V}^{\text{dipole}}, \quad \hat{V}^{\text{dipole}} = \sum_{s \neq s'} \sum_{i,j=1}^3 \mu_{ij}^{(ss')} \hat{p}_i^{(s)} \hat{p}_j^{(s')}, \\ \mu_{ij}^{(ss')} &= \frac{\delta_{ij} - 3n_i n_j}{8\pi\epsilon |\vec{x}_s - \vec{x}_{s'}|^3}, \end{aligned} \quad (\text{D.2})$$

where \hat{V} is the non-harmonic stress–stress many-body interaction we mentioned before, and \hat{V}^{dipole} is the electric dipole-dipole interaction. \vec{n} is the unit vector of $\vec{x}_s - \vec{x}_{s'}$.

We then apply a weak external phonon field $\vec{u}(\vec{x}, t)$. It alters the coefficient $\mu_{ij}^{(ss')}$ of dipole-dipole interaction, by changing the inter-dipolar positions: $\vec{x}_s - \vec{x}_{s'} \rightarrow \vec{x}_s + \vec{u}(\vec{x}_s, t) - \vec{x}_{s'} - \vec{u}(\vec{x}_{s'}, t)$:

$$\begin{aligned} \Delta\mu_{ij}^{(ss')} &= \frac{3|\vec{u}(\vec{x}_s, t) - \vec{u}(\vec{x}_{s'}, t)|}{8\pi\epsilon |\vec{x}_s - \vec{x}_{s'}|^4} \left[- (n_j m_i + n_i m_j) \right. \\ &\quad \left. + (5n_i n_j - \delta_{ij}) \frac{(\vec{u}(\vec{x}_s, t) - \vec{u}(\vec{x}_{s'}, t)) \cdot (\vec{x}_s - \vec{x}_{s'})}{|\vec{u}(\vec{x}_s, t) - \vec{u}(\vec{x}_{s'}, t)| \cdot |\vec{x}_s - \vec{x}_{s'}|} \right], \end{aligned} \quad (\text{D.3})$$

where $\vec{m} = (\vec{u}(\vec{x}_s, t) - \vec{u}(\vec{x}_{s'}, t)) / |\vec{u}(\vec{x}_s, t) - \vec{u}(\vec{x}_{s'}, t)|$ is the unit vector of $\vec{u}(\vec{x}_s, t) - \vec{u}(\vec{x}_{s'}, t)$. External phonon alters dipole moments $\hat{p}^{(s)}$ as well by changing the relative positions of positive-negative charges:

$$\Delta\hat{p}_i(\vec{x}, t) = \sum_k \frac{\partial u_i(\vec{x}, t)}{\partial x_k} \hat{p}_k(\vec{x}). \quad (\text{D.4})$$

In summary, external phonon introduces time-dependent perturbation

$$\hat{H}'_{\text{dipole}}(t) = \sum_{s \neq s'} \sum_{i,j=1}^3 \left(\Delta\mu_{ij}^{(ss')}(t) \hat{p}_i^{(s)} \hat{p}_j^{(s')} + 2\mu_{ij}^{(ss')} \Delta\hat{p}_i^{(s)}(t) \hat{p}_j^{(s')} \right). \quad (\text{D.5})$$

Now we are able to calculate phonon energy absorption with this time-dependent perturbation $H'_{\text{dipole}}(t)$,

$$\begin{aligned} \frac{dE_t^{\text{dipole}}}{dt} &= \frac{94A^2 k^2 N_0^3 \ln N_0}{960\pi^2 \epsilon^2} (1 - e^{-\beta\omega}) \omega \int \text{Im } \tilde{\chi}(\Omega) \text{Im } \tilde{\chi}(\omega - \Omega) d\Omega \\ \frac{dE_t^{\text{dipole}}}{dt} &= \frac{53A^2 k^2 N_0^3 \ln N_0}{960\pi^2 \epsilon^2} (1 - e^{-\beta\omega}) \omega \int \text{Im } \tilde{\chi}(\Omega) \text{Im } \tilde{\chi}(\omega - \Omega) d\Omega. \end{aligned} \quad (\text{D.6})$$

As mentioned earlier, $dE_{l,t}^{\text{dipole}}/dt$ is negligible compared to $dE_{l,t}^{(1)}/dt$ given by equation (3.4). To prove this, we qualitatively calculate the ratio between them:

$$\frac{dE_{l,t}^{\text{dipole}}/L_i^3 dt}{dE_{l,t}^{(1)}(L)/L^3 dt} \approx \frac{\rho c_{l,t}^2 (\text{Im } \tilde{\chi})^2 \omega_c}{L_i^3 \epsilon^2 (\text{Im } \tilde{\chi}_t)^2 \ln(\omega_c/\omega)}, \quad (\text{D.7})$$

where in the above result we assume that dielectric susceptibility $\text{Im } \tilde{\chi}$ is roughly independent of frequency up to the upper cut-off frequency ω_c . There is a critical length scale L'_c , below which $dE_{l,t}^{\text{dipole}}/dt$ is greater than $dE_{l,t}^{(1)}/dt$. To estimate the maximal possible value of L'_c , we let ω_c to take an extremely high value 10^{15} rad/s, which corresponds to an unreasonably high temperature $T_c = (\hbar\omega_c/k_B) \sim 10^4$ K. Even though we allow ω_c to take such large value, L'_c is still much smaller than $L_1 \sim 50$ Å, as shown below:

$$L'_c = \left(\frac{(\text{Im } \tilde{\chi})^2 \rho c_{l,t}^2 \hbar \omega_c}{\epsilon^2 (\text{Im } \tilde{\chi}_t)^2 \ln(\omega_c/\omega)} \right)^{\frac{1}{3}} \approx 10 \text{ Å} \ll L_1 = 50 \text{ Å}. \quad (\text{D.8})$$

The above result is obtained through the experimental data by Hunklinger and Schickfus [39]. It indicates that throughout the entire renormalization procedure, the contribution of phonon energy absorption from electric dipole-dipole interaction \hat{V}^{dipole} is always negligible, compared to that from non-harmonic stress–stress interaction \hat{V} .

One may ask the question that both \hat{V} and \hat{V}^{dipole} are $1/r^3$ long range interactions, but why are their contributions to phonon energy absorption so different? The main reason is that stress tensor operators \hat{T}_{ij} directly couple to phonon strain $\mathbf{e}(\vec{x}, t)$ via stress-strain coupling $e_{ij} \hat{T}_{ij}$, while there is no such dipole-strain coupling that directly couples electric dipoles to phonon strain field. As we have emphasized, $dE_{l,t}^{(1)}/dt$ in equation (3.4) is the most significant contribution to phonon energy absorption. It comes from the first order expansion in terms of \hat{V} and second order expansion in terms of $e_{ij} \hat{T}_{ij}$. There is no such contribution from \hat{V}^{dipole} and therefore all other contributions from it are negligible compared to $dE_{l,t}^{(1)}/dt$.

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