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UNIVERSAL PROPERTIES OF LOW-TEMPERATURE GLASS

BY

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DISSERTATION

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# Abstract

Amorphous solids (glasses) present universal properties strikingly different from that of crystalline counterparts at low temperatures, regardless of their microscopic nature. Tunneling-two-level-system model (TTLS model) successfully explained several universalities below 1K, but it cannot explain the other glass low-temperature universal properties. Based on virtual phonon exchange interaction, we develop a glass generic coupled block model to discuss two universal properties: sound velocity/dielectric constant shift, and low-temperature mechanical avalanche problem. We also successfully explain the universal property of glass Meissner-Berret ratio by using our generic coupled block model.

*Every challenging work, needs self efforts as well as guidance of elders especially those who were very close to our heart. My humble effort I dedicate to my sweet and loving*

***Father & Mother,***

*Whose affection, love, encouragement and prays of day and night make me able to get such success and honor,*

*Along with all hard working and respected*

***Teachers***

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# List of Symbols and Abbreviations

TTLS Tunneling-Two-Level-System

MB ratio Meissner-Berret ratio

$t, l$  Longitudinal, transverse

$T$  Temperature

$\beta$   $\beta = (k_B T)^{-1}$

$\rho$  Glass mass density

$m$  Glass elementary block mass

$c_{l,t}$  Longitudinal, transverse sound velocity

$\alpha$   $\alpha = 1 - c_t^2/c_l^2$

$\vec{u}(\vec{x})$  Displacement of particle at certain position  $\vec{x}$

$\vec{x}$  Position coordinate

$\vec{p}$  Momentum of certain particle

$\vec{A}$  Phonon wave amplitude

$\vec{a}$  Unit vector of phonon wave amplitude

$\vec{k}$  Phonon wave vector

$\vec{\kappa}$  Unit vector of phonon wave vector

$\omega_{k,(l,t)}$  Phonon frequency with wave number  $k$  for longitudinal/transverse phonon

$e_{ij}$  Phonon strain field

$\hat{T}_{ij}$  Stress tensor

$\chi_{ijkl}$  Stress tensor susceptibility

$\chi_{l,t}$  Longitudinal/transverse component of stress tensor susceptibility

$\gamma_{l,t}$  TTLS-phonon coupling constant for longitudinal and transverse phonon

$\gamma_l/\gamma_t$  Meissner-Berret ratio

$\vec{x}_s$  Location of the  $s$ -th glass unit block

$\vec{x}_{ss'}$   $\vec{x}_s - \vec{x}_{s'}$

$\vec{n}_{ss'}$  Unit vector of  $\vec{x}_{ss'}$

$\Lambda_{ijkl}^{(ss')}$  Coefficient of virtual phonon exchange interaction



# Chapter 1

## Introduction

It has been more than 50 years since the first experiment[1] by Zeller and Pohl showed at ultra-low temperatures below 1K the thermal and acoustic properties of amorphous solids (glasses) behave entirely different from that of crystalline counterparts. In 1972, Anderson, Halperin and Varma[3] group and Phillips[19] independently developed a microscopic phenomenological low-temperature glass model which was later known as tunneling-two-level-system model (TTLS). The effective Hamiltonian for TTLS model is the summation of elastic (phonon) part of Hamiltonian, a set of two level systems randomly embedded in glass material, and the coupling between two-level-system and phonon strain field.

$$\hat{H} = \hat{H}_{\text{ph}} + \frac{1}{2} \begin{pmatrix} E & 0 \\ 0 & -E \end{pmatrix} + \frac{\gamma_{l,t}}{2} \begin{pmatrix} \Delta/E & \Delta_0/E \\ \Delta_0/E & -\Delta/E \end{pmatrix} \mathbf{e}(t) \quad (1.1)$$

where  $E = \sqrt{\Delta^2 + \Delta_0^2}$ . The coupling constants between longitudinal/transverse phonon strain and TLS are denoted as  $\gamma_{l,t}$ , which are adjustable parameters. Together with random distributions of TTLS model parameters[10], it not only explained existing experiments successfully, such as linear temperature dependence of heat capacity, but also predicted new experiments such as phonon echo[38] and saturation[36] phenomena. In chapter 1, we will discuss the significance of TTLS model in details.

However, TTLS model has a number of problems. First, while TTLS successfully explained several universal properties of amorphous solid below 1K, there are more universalities cannot be explained for temperatures around  $1\text{K} < T < 50\text{K}$ [25], e.g. universal thermal conductivity plateau around 10K, and universal internal friction  $Q^{-1}$  plateau between  $10\text{K} < T < 50\text{K}$ [34]. Second, the model itself has too many adjustable parameters, for example, random distribution function  $f(E, \Delta)$  for the diagonal and off-diagonal matrix elements  $E, \Delta$  of two level system; coupling constants  $\gamma_{l,t}$ , etc. Experimental results could always be explained by adjusting these parameters within a certain range. Third, the model lacks the consideration that as the interaction with phonon strain field, TTLS must generate a mutual RKKY-type interaction[42]. Taking this virtual-phonon exchange interaction into account may not only change current theoretical results, but also question the validity of TTLS.

Besides the above general TTLS problems, in this thesis we will focus on 3 specific universal properties which cannot be fully explained within TTLS model. They are: universal shifts on sound velocity and dielectric constant, universal Meissner-Berret ratio and glass mechanical avalanche phenomena. We want to focus on these universal acoustic and mechanical properties by developing a theory of coupled generic blocks. We start by expanding non-elastic part of glass Hamiltonian in orders of strain field  $e_{ij}(\vec{x})$  and derive the non-elastic stress-stress susceptibility via linear response theory. By putting in many-body interaction generated from virtual phonon exchange process, we set up the renormalization recursion relation for non-elastic susceptibilities at various length scales. Our goal is to prove that glass universal properties essentially come from many-body interaction, independent of materials' microscopic structure and chemical compound.

The first problem we will discuss in this thesis is the glass universal shift on sound velocity and dielectric constant. To verify the existence of two-level-systems, L. Piché, R. Maynard, S. Hunklinger and J. Jäckle[33] studied two-level-systems' influence on the variation of longitudinal sound velocity in vitreous silica Suprasil I at temperatures  $0.28\text{K} < T < 4.2\text{K}$  and frequencies  $30\text{MHz} < f < 150\text{MHz}$ . The sound velocity shift was found to be logarithmically dependent on temperature. At high frequency low temperature resonance regime with  $\omega\tau \gg 1$  ( $\tau$  is the effective thermal relaxation time, please refer to chapter 3 for detailed discussions) the sound velocity increases with increasing temperature. This sound velocity shift in resonance regime is independent of phonon frequency. At low frequency high temperature relaxation regime with  $\omega\tau \ll 1$  the velocity decreases with increasing temperature. Such sound velocity increase-decrease transition occurs at the transition point  $\omega\tau(T) \approx 1$ , which means the transition temperature  $T$  is functional of phonon frequency. However, as long as the sound velocity measurement enters into relaxation regime, it turns out to be frequency independent as well. In the rest of this thesis we will discuss the temperature dependence of sound velocity in relaxation and resonance regimes separately, so we assume that sound velocity shift is frequency independent in both relaxation and resonance regimes. Such universality has been observed in amorphous materials such as vitreous silica, lithium-doped KCl[4] and silica based microscopic cover glass[8], etc.. By averaging over random parameters of glass two-level-system susceptibility, TTLS model successfully explained the logarithmic temperature dependence of sound velocity shift[19, 10]. It also proves that the slope of  $\ln T$  dependence is negative in relaxation regime and positive in resonance regime. The sound velocity slope ratio between relaxation and resonance regimes is  $\mathcal{C}^{\text{rel}} : \mathcal{C}^{\text{res}} = -1/2 : 1$ , which agrees quite well with silica based microscopic cover glass measurements[8]. However at least to the author's knowledge, it is the only amorphous material with the absolute value of slope in relaxation regime smaller than that of resonance regime. Other materials, however, present the absolute value of slope in relaxation regime equal or

slightly greater than that of resonance regime: vitreous silica Suprasil I[33], PdSiCu[9], Zr-Nb[17], lithium-doped KCl[4], vitreous silica[11], metallic glass[13]  $\text{Ni}_{81}\text{P}_{19}$ , etc. (the electron-TTLS coupling in metallic glass is relatively weak compared to phonon-TTLS coupling, so conducting electrons are not strong enough to affect sound velocity[10]). S. Hunklinger and C. Enss[12] suggest that most of the sound velocity slope ratios of glass materials are rather  $-1$  to  $1$ , probably due to the interaction between tunneling systems, because glass defects are highly concentrated. Our purpose is to set up a generic glass model to prove such universal slope ratio of temperature dependence on sound velocity shift, in relaxation and resonance regimes. We hope our renormalization technique would lead to the universal shift of sound velocity, but right now the renormalization equations in chapter 4 lead to the increasing behavior of relaxation and resonance susceptibilities rather than the expected decreasing behavior as the length scale increases. Moreover, the fixed point in Eqs.(4.19),  $\chi^{\text{rel}} = -2\chi^{\text{res}}(\omega = 0)$  can never be reached, due to the fact that both of relaxation and zero-frequency resonance susceptibilities are negative — they will always have the same sign throughout the entire renormalization process. It is at this point that our renormalization technique cannot explain the universal sound velocity shift.

By assuming that electric field couples to two-level-systems[7], the calculation of TTLS model on dielectric constant shift is similar with sound velocity shift, but the dielectric shift slope ratio between relaxation and resonance regimes is  $\mathcal{C}^{\text{rel}} : \mathcal{C}^{\text{res}} = +1/2 : -1$ . However, dielectric measurements on various amorphous materials such as vitreous silica Suprasil W and vitreous  $\text{As}_2\text{S}_3$ [14], vitreous silica Suprasil I[6] and borosilicate glass (BK7)[15] indicate that the slope ratio is  $\mathcal{C}^{\text{rel}} : \mathcal{C}^{\text{res}} = +1 : -1$ , regardless of their microscopic nature. In this thesis we also try to use electric dipole-dipole interaction to carry out universal glass dielectric constant shift. However, our model and renormalization procedure cannot prove this universal property as well, because of the same reason as universal sound velocity shift, that the relaxation and resonance susceptibilities have the same sign, and the fixed point  $\chi^{\text{rel}} = -2\chi^{\text{res}}(\omega = 0)$  can never be reached.

The second goal of this thesis is to use our generic coupled block model to understand the mechanical avalanche behavior of three-dimensional insulating glass. The reader should be aware that it is the first time to apply our model in glass mechanical avalanche problem. Therefore our purpose is not to solve the entire glass avalanche problem from microscopic point of view; instead we want to provide some first-step results for future people to continue studying this problem. We consider a block of amorphous material under the deformation of static, uniform strain. With the slowly increasing strain the bulk glass behaves elastically until it reaches critical strain value. After that the stress ( $\mathbf{T}$ ) suddenly drops to a lower value. A more convenient quantity is the mechanical stress-stress susceptibility  $\chi_{ijkl} = \delta T_{ij} / \delta e_{kl}$ . At critical strain field when irreversible process happens, stress-stress susceptibility presents an abrupt positive-negative transition

when strain field passes through critical value. Our main goal is to prove the existence of such positive-negative transition in glass mechanical susceptibility.

We successfully explained the third universal property in this thesis, the glass universal Meissner-Berret ratio. In 1987, Meissner and Berret[45] measured 18 different kinds of glass materials' coupling constants  $\gamma_{l,t}$ . They pointed out that the coupling constants  $\gamma_{l,t}$  are not arbitrary: below temperature  $T < 1\text{K}$ , their ratio  $\gamma_l/\gamma_t$  turns out to lie between 1.44 and 1.84 for a wide variety of amorphous materials, regardless of their chemical compound and microscopic molecular structure. Such universality cannot be explained within TTLS model since the model itself is based on the coupling constants. We believe that there must be a more general model to describe universal properties of low-temperature glass, including universal ratio  $\gamma_l/\gamma_t$ . In the rest of this thesis, we use "Meissner-Berret Ratio" to stand for "TTLS coupling constants' ratio  $\gamma_l/\gamma_t$ ". We consider this problem by calculating glass resonance phonon energy absorption due to the input of external longitudinal and transverse phonons. Within TTLS model the resonance energy absorption per unit time  $\dot{E}_{l,t}$  is proportional to the square of coupling constant  $\gamma_{l,t}$ ; in our generic coupled block model the resonance energy absorption is proportional to the imaginary part of resonance susceptibility, and it is only functional of longitudinal/transverse sound velocity ratio. This experimentally measurable quantity does not rely on adjustable parameters. We believe our theory can help explain the universality of Meissner-Berret Ratio.

The organization of this thesis is as follows: in chapter 2 we give a short review of the traditional model – tunneling-two-level-system model, the contributions of it to glass low-temperature behavior explanations, and limitations of it. In chapter 3 we set up our generic coupled block model and non-elastic stress-stress interaction via virtual phonon exchange process, with the presence of external phonon field. We also introduce the most important concepts, elastic and non-elastic stress-stress susceptibilities. In chapter 4 we study glass universal shift on sound velocity and dielectric constant. We study real space renormalization recursion relation between small and large length scale non-elastic stress-stress susceptibilities. In chapter 5 we work on the microscopic explanation of glass mechanical avalanche phenomena. We derive the recursion relation between small and large length scale static susceptibilities. In chapter 6 we explore the universal Meissner-Berret ratio in glass. We calculate the resonance phonon energy absorption of a group of interacting single blocks due to the input of external longitudinal (transverse) phonon strain field. By assuming the external strain field is weak enough that the stress-strain coupling can be treated as perturbation, we expand resonant phonon energy absorption up to the second order of coupling, and derive resonance energy absorption recursion relation between single block and super block. We use such real space renormalization procedure to carry out the Meissner-Berret ratio at experimental length scale. We prove this experimental

measurable quantity is independent of the material's microscopic nature. We also give a detailed discussion on the influence of electric dipole interaction on Meissner-Berret ratio for dielectric amorphous solids. The influence of electric dipole-dipole interaction to Meissner-Berret ratio is negligible. In the appendix (A) we give a detailed derivation on non-elastic stress-stress interaction coefficient  $\Lambda_{ijkl}^{(ss')}$ , and point out 4 differences between our result and that derived by Joffrin and Levelut[42].

## Chapter 2

# Tunneling-Two-Level-System Model

At low temperatures the only excitations to contribute the specific heat of insulating crystals are long wavelength phonon modes. For  $T < 1\text{K}$ , much smaller than Debye temperature, the specific heat of insulating crystal has a  $T^3$ -dependence, for example, the specific heat of crystalline quartz is  $C = 0.55T^3 \mu/\text{gK}$ [1]. However, the specific heat of glass is considerably higher. If we subtract the phonon contribution  $C_D$  calculated from Debye's theory from the glass specific heat  $C$ , the excess glass specific heat  $C_a = C - C_D$  is characteristic of the amorphous state. The additional specific heat capacity can be approximated by

$$C_a = a_1 T^{1+\delta} + a_3 T^3 \quad (2.1)$$

for example, the exponents are  $\delta = 0.22$  for Suprasil and  $\delta = 0.3$  for Suprasil W[8]. The glass excess heat

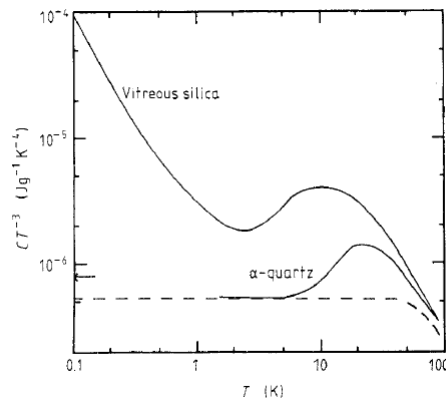


Figure 2.1: The heat capacity comparison between amorphous material (vitreous silica) and the crystalline version (quartz) of it by Zeller and Pohl[1].

capacity  $C_a$  at 0.1 K is about two orders of magnitude greater in the glass than in the crystal. Besides the anomalous specific heat, glass thermal conductivity also differs from that of crystalline solids. The thermal conductivity can be interpreted qualitatively by kinetic formula

$$\kappa = \frac{1}{3} C v_s l \quad (2.2)$$

where  $l$  is the phonon mean free path. At low temperatures in crystals phonons are scattered by defects in the crystal or by the surfaces of the sample, so that  $l$  is independent of temperature and  $\kappa$  is therefore proportional to  $T^3$ . However in glass below 1K  $\kappa$  increases quadratically as the increase of temperature and then enters into a thermal conductivity plateau with temperatures  $4\text{K} < T < 20\text{K}$ . Similar results are seen in a wide range of other amorphous solids; oxide, chalcogenide, elemental, polymeric and metallic glasses all present the same behaviour. The universality of the temperature dependences of glass heat capacity proportional to  $T$  and thermal conductivity proportional to  $T^2$  provide great attractions for theorists. Since

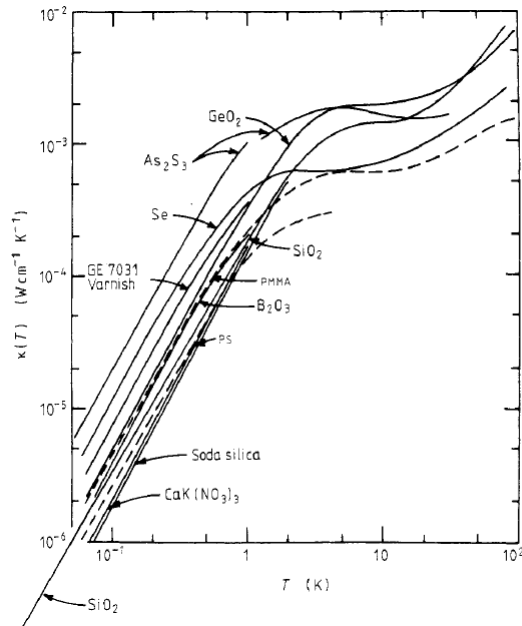


Figure 2.2: The thermal conductivity for different amorphous materials by R. B. Stephens[48]. At low temperatures below 1K, it increases quadratically as the increase of temperature. Between 4K and 20K it enters in a universal plateau, regardless of the materials' microscopic structure and chemical compound.

the anomalous properties of glass are observed down to very low temperatures, we want to develop an effective model to describe such low-temperature behaviors. In the regular lattice of a crystal all atoms or molecules occupy a well defined position, allowing only one possible configuration. In glass the random structure of glass material can be realized as a large number of different configurations. Therefore we assume there are a group of tunneling-two-level-systems randomly embedded in glass material. They can occupy at least two different positions or configurations. We may introduce "particles" of unknown microscopic mechanism moving in double-well potentials. In each of the wells such particles move between these wells[7], and they have a series of vibrational states separated by an energy  $\hbar\Omega$  which is of the order of the Debye energy. At low-temperatures we are only interested in the ground states with the wave functions  $\psi_L$  and  $\psi_R$  for the particles located either in the "left" or "right" well, respectively, in the following figure. The

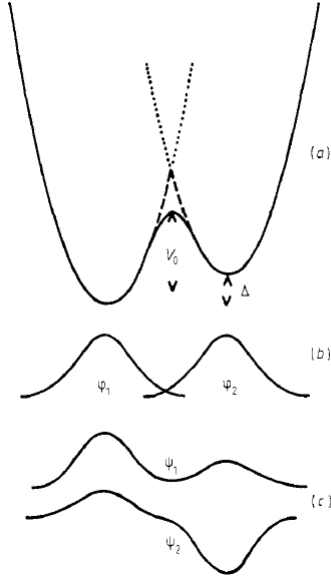


Figure 2.3: Double well potential by Phillips[19].

difference of double well potential minima is referred to as the “asymmetry”  $\Delta$ . The tunneling strength between two potentials is  $\Delta_0 = \hbar\Omega e^{-\lambda}$ . The tunneling parameter  $\lambda = d(2mV/\hbar^2)^{1/2}$  represents the overlap of the wave function  $\psi_L$  and  $\psi_R$ .  $d$  is the separation between the two wells,  $m$  the effective mass of the tunneling particles and  $V$  the barrier between two minima. In the basis  $(\psi_L, \psi_R)$  the Hamiltonian of a single tunneling system is given by [37]

$$\hat{H}_{\text{TLS}} = \frac{1}{2} \begin{pmatrix} \Delta & \Delta_0 \\ \Delta_0 & -\Delta \end{pmatrix} \quad (2.3)$$

Anderson, Halperin, Varma and Phillips assume that the TTLS parameters  $\Delta$  and  $\lambda$  are independent of each other and to have the distribution function as follows,

$$P(\Delta, \lambda)d\Delta d\lambda = \bar{P}d\Delta d\lambda \quad (2.4)$$

which means the distribution function  $P(\Delta, \lambda)$  is uniform. Because of the exponential dependence of  $\Delta_0$  on  $\lambda$ , only a relatively small range of  $\lambda$  is responsible for a large range of  $\Delta_0$  and over this limited range the distribution of  $\lambda$  can be taken as a constant. Therefore the distribution function for  $\Delta$  and  $\Delta_0$  is given by

$$f(\Delta, \Delta_0) = \frac{\bar{P}}{\Delta_0} \quad (2.5)$$



Tunneling systems couple to their environments via strain and electric fields. Since both mechanisms can be described in the same way, we will only discuss the coupling to strain fields. The coupling can be realized by transitions from one well to the other. This phonon assisted tunneling process leads to a change of  $\Delta_0$  and  $\Delta$ . These “diagonal and off-diagonal matrix elements variation” can be described by deformation potentials  $\gamma_{\Delta_0} = \delta\Delta_0/2\delta e$  and  $\gamma_{\Delta} = \delta\Delta/2\delta e$ . Thus the coupling between tunneling system and phonon strain field  $\mathbf{e}(t)$  can be written as

$$\hat{H}_{\text{coup}} = \begin{pmatrix} \gamma_{\Delta}^{l,t} & \gamma_{\Delta_0}^{l,t} \\ \gamma_{\Delta_0}^{l,t} & -\gamma_{\Delta}^{l,t} \end{pmatrix} \mathbf{e}(t) \quad (2.6)$$

where  $l$  and  $t$  denotes the coupling with longitudinal/transverse phonon strain fields. Usually we assume that  $\gamma_{\Delta} \gg \gamma_{\Delta_0}$ , which means the strain fields mainly couple to the asymmetry  $\Delta$ . At the first glance this assumption is unusual. However coupling constants associated with the variation of the geometry are expected to be rather small, namely of the order the energy splitting itself[5, 10, 19]. Based the assumption that TTLS-phonon coupling term is diagonal, we would like to rewrite glass TTLS Hamiltonian in the basis of TTLS energy eigenvalues  $\pm E = \pm\sqrt{\Delta^2 + \Delta_0^2}$ :

$$\hat{H} = \hat{H}_{\text{ph}} + \frac{1}{2} \begin{pmatrix} E & 0 \\ 0 & -E \end{pmatrix} + \frac{\gamma_{l,t}}{2} \begin{pmatrix} \Delta/E & \Delta_0/E \\ \Delta_0/E & -\Delta/E \end{pmatrix} \mathbf{e}(t) \quad (2.7)$$

where the coupling constants  $\gamma_{l,t}$  are for TTLS-longitudinal/transverse phonon couplings. The diagonal matrix element of TTLS-phonon coupling represents the TTLS energy eigenvalue shift due to the external perturbation, and it is further represented by relaxation process, while the off-diagonal matrix element of coupling stands for the transitions between TTLS different eigenstates, and it is further represented by resonance process. Based on the above TTLS model assumptions, in this chapter we give a short review of TTLS model calculations on the 3 universal properties we will talk about in later chapters.

Consider the external phonon field which makes the transitions between TTLS state 1 (with energy eigenvalue  $+E$ ) and 2 (with  $-E$ ). If we denote the transition probability rate from state 1 to state 2 as  $\omega_{12}$ , and  $\omega_{21}$  for state 2 to 1, then the time derivative of state 1 and 2 probability  $\dot{P}_1, \dot{P}_2$  obey the following equation of motion:

$$\dot{P}_1 = -P_1\omega_{12} + P_2\omega_{21} \quad \dot{P}_2 = P_1\omega_{12} - P_2\omega_{21} \quad \Rightarrow \quad \dot{P}_1 = -P_1(\omega_{12} + \omega_{21}) + \omega_{21} \quad (2.8)$$

In thermal equilibrium we have  $P_1\omega_{12} = P_2\omega_{21}$ . The relaxation time of two-level-system  $\tau$  is defined by

$\dot{P}_1 = -P_1/\tau$ , and is further given by  $\tau^{-1} = \omega_{12}(1 + e^{\beta E})$ . The transition rate  $\omega_{12}$  can be calculated by Fermi golden rule,

$$\omega_{12} = \sum_{\alpha=l,t} \frac{2\pi}{\hbar} |\langle \psi_1 | \hat{H}_{\text{int}} | \psi_2 \rangle|^2 \frac{g(E)}{e^{\beta E} - 1} \quad (2.9)$$

where  $\hat{H}_{\text{coup}}$  is the coupling between phonon strain field and two-level-systems,  $\alpha$  is phonon polarization,  $g(E)$  is phonon density of states. Therefore one gets the relaxation time of an TLS with  $(\Delta, \Delta_0)$

$$\tau^{-1}(E) = \sum_{\alpha} \frac{\gamma_{\alpha}^2}{c_{\alpha}^5} \frac{E \Delta_0^2}{2\pi \rho \hbar^4} \coth\left(\frac{1}{2}\beta E\right) \quad (2.10)$$

where  $\Delta_0$  comes from the off-diagonal matrix element of TTLS-phonon coupling. The phonon absorption process corresponds to phonon number reduction:  $g(E)\dot{n}_{\text{ph}}(E) = -\dot{P}_1$ . Plugging in Eq.(2.8) one obtains the phonon scattering rate  $\tau_{\text{ph}}^{-1}$

$$\tau_{\alpha,\text{ph}}^{-1}(\Delta_0, \Delta) = \frac{2\pi}{\hbar} |\langle \psi_1 | \hat{H}_{\text{int}}(\Delta_0, \Delta) | \psi_2 \rangle|^2 \tanh\left(\frac{1}{2}\beta E\right) = \frac{\pi \gamma_{\alpha}^2 \omega \Delta_0^2}{\rho c_{\alpha}^3 E^2} \tanh\left(\frac{1}{2}\beta E\right) \quad (2.11)$$

where  $E = \sqrt{\Delta^2 + \Delta_0^2}$ . Since the total phonon scattering process comes from all the two-level-systems with different parameters  $\Delta, \Delta_0$ , and  $\sqrt{\Delta^2 + \Delta_0^2} = E$ , one needs to sum over all two-level-systems with different  $\Delta_0$  ranges from  $-E$  to  $E$  to obtain mean free path.

$$l_{\alpha}^{-1}(\omega) = (c_{\alpha} \tau_{\alpha,\text{ph}})^{-1} = \int_{-E}^E f(\Delta, \Delta_0) c_{\alpha} \tau_{\alpha,\text{ph}}^{-1}(\Delta_0) d\Delta_0 = \frac{\bar{P} \pi \gamma_{\alpha}^2 \omega}{\rho c_{\alpha}^3} \tanh\left(\frac{1}{2}\beta \hbar \omega\right) \quad (2.12)$$

where the uniform distribution probability  $\bar{P}$  is given by Eq.(2.4). Given the phonon mean free path  $l_{\alpha}$  with polarization  $\alpha$ , the sound velocity shift as the function of temperature can be calculated from the real part of response function, and it is further given by Kramers-Kronig relation as

$$c_{\alpha}(\omega, T) - c_{\alpha}(\omega, 0) = \Delta c_{\alpha}(\omega) = \frac{1}{\pi} \text{P} \int_0^{\infty} \frac{c_{\alpha}^2 l_{\alpha}^{-1}(\Omega)}{\omega^2 - \Omega^2} d\Omega \quad (2.13)$$

where the integral is principle value. Using the form of mean free path Eq.(2.12) the sound velocity shift is given by

$$\left. \frac{\Delta c_{\alpha}}{c_{\alpha}} \right|_{\text{res}} = \frac{\bar{P} \gamma_{\alpha}}{\rho c_{\alpha}^2} \ln\left(\frac{T}{T_0}\right) \quad (2.14)$$

This result is the main tool to experimentally measure the values of  $\bar{P} \gamma_{\alpha}^2$ . Please note that the previous

calculations are from phonon absorption/emission process, so Eq.(2.14) is just resonance process sound velocity shift. However in high temperature low frequency relaxation regime, both of resonance and relaxation processes contribute to the sound velocity shift. The relaxation susceptibility can be obtained by introducing thermal relaxation time  $\tau$  for two-level-system. Consider the population difference  $\Delta P = P_1 - P_2$  between two levels, relaxation process gives

$$\frac{\partial \Delta P}{\partial t} = -\frac{\Delta P - \Delta P^{\text{ins}}}{\tau} \quad (2.15)$$

where Eq.(2.15) is the equation of instantaneous relaxation of  $e(t)$ , and  $\Delta P^{\text{ins}}$  is the instantaneous distribution function for the population difference. The expectation value of the two-level-system Hamiltonian can be perturbatively expanded in orders of external strain field. The susceptibility is defined as the first order derivative of the expectation value with respect to strain field:

$$\chi(\omega) = \frac{\delta \langle \hat{H}_{\text{TLS}} \rangle}{\delta e(\omega)} = \frac{\chi(0)}{1 - i\omega\tau} \quad (2.16)$$

with

$$\chi(0) = \frac{\gamma_\alpha^2 \Delta^2}{4k_B T E^2} \text{sech}^2 \left( \frac{1}{2} \beta E \right) \quad (2.17)$$

in relaxation regime, both of relaxation and resonance susceptibilities contribute to the sound velocity shift. The relaxation part contribution is

$$\left. \frac{\Delta c_\alpha}{c_\alpha} \right|_{\text{rel}} = \frac{\text{Re } \chi(\omega)}{2\rho c_\alpha^2} \quad (2.18)$$

We need to take all possible two-level-systems that contribute to the relaxation process, which is, to sum over all possible TTLS parameters  $\Delta$  and  $\Delta_0$ . A more convenient way to calculate this summation is to transform  $\Delta, \Delta_0$  summation into  $E, \tau$  summation, where  $\tau$  is the relaxation time for a certain TTLS. From Eq.(2.10), the two-level-system relaxation time is inversely proportional to  $\Delta_0^2$ . Therefore the smallest possible relaxation time  $\tau$  is obtained by setting  $\Delta_0 = E$ , which gives

$$\tau_{\min} = \left[ \sum_\alpha \frac{\gamma_\alpha^2}{c_\alpha^5} \frac{E^3}{2\pi\rho\hbar^4} \coth \left( \frac{1}{2} \beta E \right) \right]^{-1} \Rightarrow \frac{\Delta^2}{E^2} = 1 - \frac{\tau_{\min}}{\tau} \quad (2.19)$$

Also, it is important to note, that the minimum of relaxation time  $\tau_{\min} = T^{-3} \left[ k_B^3 \sum_\alpha \frac{\gamma_\alpha^2}{c_\alpha^5} \frac{(\beta E)^3}{2\pi\rho\hbar^4} \coth \left( \frac{1}{2} \beta E \right) \right]^{-1}$ .

Therefore, the  $\Delta, \Delta_0$  distribution function,  $f(\Delta, \Delta_0)$  can be transformed to  $g(E, \tau)$  distribution function via Jacobian determinant:

$$g(E, \tau)dEd\tau = f(\Delta, \Delta_0)\frac{\Delta_0 E}{2\tau\Delta}d\Delta d\Delta_0 \quad (2.20)$$

Plugging in the specific form we made for TTLS model, that  $f(\Delta, \Delta_0) = \bar{P}/\Delta_0$ , we have the distribution function  $g(E, \tau)$  given by

$$g(E, \tau) = -\frac{\bar{P}}{2\tau(1 - \tau_{\min}(E)/\tau)^{1/2}} \quad (2.21)$$

Finally, the summation over different configurations of two-level-systems which contribution to the sound velocity shift for relaxation susceptibility is

$$\begin{aligned} \left. \frac{\Delta c_\alpha}{c_\alpha} \right|_{\text{rel}} &= \frac{\gamma_\alpha^2}{2\rho c_\alpha^2 k_B T} \int_0^\infty dE \int_{\tau_{\min}}^\infty \text{sech}^2\left(\frac{E}{2k_B T}\right) \left(1 - \frac{\tau_{\min}}{\tau}\right) \frac{g(E, \tau)}{1 + \omega^2 \tau^2} d\tau \\ &= \frac{\gamma_\alpha^2}{2\rho c_\alpha^2 k_B T} \int_0^\infty dE \int_{\tau_{\min}}^\infty \text{sech}^2\left(\frac{E}{2k_B T}\right) \left(1 - \frac{\tau_{\min}}{\tau}\right) \frac{1}{1 + \omega^2 \tau^2} \left[ -\frac{\bar{P}}{2\tau(1 - \tau_{\min}/\tau)^{1/2}} \right] d\tau \\ &= -\frac{\bar{P}\gamma_\alpha^2}{2\rho c_\alpha^2} \int_0^\infty \text{sech}^2\left(\frac{E}{2k_B T}\right) d\left(\frac{E}{2k_B T}\right) \int_{\tau_{\min}}^\infty \left(1 - \frac{\tau_{\min}}{\tau}\right)^{1/2} \frac{1}{1 + \omega^2 \tau^2} \frac{d\tau}{\tau} \\ &\approx -\frac{\bar{P}\gamma_\alpha^2}{2\rho c_\alpha^2} \int_0^\infty \text{sech}^2\left(\frac{E}{2k_B T}\right) d\left(\frac{E}{2k_B T}\right) \int_{\tau_{\min}}^\infty \frac{d\tau}{\tau} \\ &= -\frac{\bar{P}\gamma_\alpha^2}{2\rho c_\alpha^2} \int_0^\infty \text{sech}^2\left(\frac{E}{2k_B T}\right) d\left(\frac{E}{2k_B T}\right) \ln\left(\frac{\tau_{\text{cutoff}}}{\tau_{\min}}\right) \\ &= -\frac{\bar{P}\gamma_\alpha^2}{2\rho c_\alpha^2} \int_0^\infty \text{sech}^2\left(\frac{E}{2k_B T}\right) d\left(\frac{E}{2k_B T}\right) \ln\left(\frac{\tau_{\text{cutoff}}}{T^{-3} \left[ k_B^3 \sum_\alpha \frac{\gamma_\alpha^2 (\beta E)^3}{c_\alpha^5 2\pi\rho\hbar^4} \coth\left(\frac{1}{2}\beta E\right) \right]^{-1}}\right) \\ &= -\frac{\bar{P}\gamma_\alpha^2}{2\rho c_\alpha^2} \int_0^\infty \text{sech}^2\left(\frac{E}{2k_B T}\right) d\left(\frac{E}{2k_B T}\right) \ln\left(\frac{\tau_{\text{cutoff}}}{T^{-3}}\right) \\ &\quad - \frac{\bar{P}\gamma_\alpha^2}{2\rho c_\alpha^2} \int_0^\infty \text{sech}^2\left(\frac{E}{2k_B T}\right) d\left(\frac{E}{2k_B T}\right) \ln\left[ k_B^3 \sum_\alpha \frac{\gamma_\alpha^2 (\beta E)^3}{c_\alpha^5 2\pi\rho\hbar^4} \coth\left(\frac{1}{2}\beta E\right) \right] \\ &= -\frac{\bar{P}\gamma_\alpha^2}{2\rho c_\alpha^2} \int_0^\infty \text{sech}^2\left(\frac{E}{2k_B T}\right) d\left(\frac{E}{2k_B T}\right) \ln(T^3) - \text{Const.} \\ &= -\frac{3\bar{P}\gamma_\alpha^2}{2\rho c_\alpha^2} \ln T - \text{Const.} = -\frac{3}{2} \frac{\bar{P}\gamma_\alpha^2}{\rho c_\alpha^2} \ln\left(\frac{T}{T_0}\right) \end{aligned} \quad (2.22)$$

The factor of 3 comes from the  $T^{-3}$  dependence of  $\tau_{\min}$  (after taking integration over variable  $E$ ). This result has the same form as that of resonance process sound velocity shift, but differs for a factor of  $-\frac{3}{2}$ . Therefore in relaxation regime the sound velocity shift is the summation of relaxation and resonance regimes,

it is further given by

$$\left. \frac{\Delta c_\alpha}{c_\alpha} \right|_{\text{tot}} = \left. \frac{\Delta c_\alpha}{c_\alpha} \right|_{\text{rel}} + \left. \frac{\Delta c_\alpha}{c_\alpha} \right|_{\text{res}} = -\frac{1}{2} \frac{\bar{P} \gamma_\alpha^2}{\rho c_\alpha^2} \ln \left( \frac{T}{T_0} \right) \quad (2.23)$$

Finally, we reach the conclusion from TTLS theory: at low-temperature (below the order of 10K), sound velocity changes as the logarithmic of temperature. It increases as the increase of temperature in resonance regime, while decreases with the increase of temperature in relaxation regime. Further more, the slope ratio between resonance and relaxation regime is  $1 : (-1/2)$ . However, as we will show in chapter 4, most of the glass materials' experiment indicate that the slope ratio is  $1 : -1$  rather than  $1 : (-1/2)$ .

From the results Eq.(2.14, 2.23), sound velocity shift slope is the function of  $\bar{P}, \gamma_\alpha, \rho$  and  $c_\alpha$ . In 1987, Meissner and Berret[45] measured 18 different kinds of glass materials' sound velocity shift slope, including organic material (PMMA), chemically pured material (a-SiO<sub>2</sub>) and chemically mixed material (BK7). They calculate  $\gamma_{l,t}$  from longitudinal and transverse sound velocity measurements based on the assumption that TTLS model is a suitable description for them. They find, that the ratio  $\gamma_l/\gamma_t$  ranges from  $1.44 \sim 1.84$  for these 18 materials, most of them are around  $1.5 \sim 1.6$ . Such universality cannot be explained within TTLS model. In chapter 6 we will discuss where does this universality come from.

Finally in chapter 5 we also give a tentative microscopic explanation regarding glass mechanical avalanche phenomena. To our knowledge there is no obvious explanation from TTLS model to solve this problem. We will discuss the mechanical failure of glass material with the presence of externa static, uniform strain with our generic coupled block model in chapter 5.

# Chapter 3

## The Generic Coupled Block Model

In this chapter we want to develop a generalized glass Hamiltonian based on a set of interacting generic blocks. We start by expanding non-elastic part of glass Hamiltonian in orders of strain field  $e_{ij}(\vec{x})$  and consider the coefficient of first order expansion with respect to strain field, namely non-elastic stress tensor. We further define non-elastic stress-stress susceptibility defined by the first order derivative of non-elastic stress tensor with respect to strain field. We also set up the many body interaction between different blocks, due to the exchange process of virtual phonons. In the following chapters 4, 5 and 6, we will discuss the universal properties of low-temperature glass from the virtual phonon exchange interaction.

### 3.1 Non-Elastic Stress-Stress Susceptibility

Let us consider a block of glass with the dimension  $L$  much greater than the atomic distance  $a \sim 10\text{\AA}$ . The elastic strain  $e_{ij}(\vec{x})$  can be defined as the spacial derivative of displacement  $\vec{u}(\vec{x})$  of the matter located at position  $\vec{x}$ :

$$e_{ij}(\vec{x}) = \frac{1}{2} \left( \frac{\partial u_i(\vec{x})}{\partial x_j} + \frac{\partial u_j(\vec{x})}{\partial x_i} \right) \quad (3.1)$$

In this section, we have not considered any external strain field yet. We write  $\hat{H}^{\text{tot}}$  for the Hamiltonian of glass, and expand it in orders of elastic intrinsic strain field  $e_{ij}$  in long wavelength limit ( $\lambda \gg a$ ):

$$\hat{H}^{\text{tot}} = \hat{H}_0^{\text{tot}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{tot}}(\vec{x}) + \mathcal{O}(e_{ij}^2) \quad (3.2)$$

where the definition of stress tensor  $\hat{T}_{ij}^{\text{tot}}(\vec{x})$  is the first order derivative of Hamiltonian with respect to intrinsic strain field

$$\hat{T}_{ij}^{\text{tot}}(\vec{x}) = \frac{\delta \hat{H}^{\text{tot}}}{\delta e_{ij}(\vec{x})} \quad (3.3)$$

Next we plug in an external infinitesimal strain field,  $e_{ij}(\vec{x}, t)$ , and measure the stress response  $\langle \hat{T}_{ij}^{\text{tot}} \rangle(\vec{x}, t)$ . We define the glass stress-stress susceptibility, and the derivative of stress response  $\langle \hat{T}_{ij}^{\text{tot}} \rangle(\vec{x}, t)$  with respect to external infinitesimal strain  $e_{kl}(\vec{x}, t)$ . The susceptibility is taken for the glass block much larger than atomic distance:

$$\chi_{ijkl}^{\text{tot}}(\vec{x} - \vec{x}'; t - t') = \frac{\delta \langle \hat{T}_{ij}^{\text{tot}} \rangle(\vec{x}, t)}{\delta e_{kl}(\vec{x}', t')} \quad (3.4)$$

In this definition of susceptibility Eq.(3.4) the average operator  $\langle \rangle$  represents thermal average and quantum average: for an arbitrary operator  $\hat{A}$  (here the operator is the stress tensor operator  $\hat{T}_{ij}^{\text{tot}}(\vec{x})$ ),  $\langle \hat{A} \rangle = \sum_m \mathcal{Z}^{-1} e^{-\beta E_m} \langle m, t | \hat{A} | m, t \rangle$  with  $|m\rangle$  the eigenbasis of Hamiltonian  $\hat{H}_0^{\text{tot}}$  and  $\mathcal{Z}$  the partition function with temperature  $\beta = (k_B T)^{-1}$ . Susceptibility is functional of temperature, but for notation simplicity we write  $\chi^{\text{tot}}(\vec{x} - \vec{x}'; t - t'; T)$  as  $\chi^{\text{tot}}(\vec{x} - \vec{x}'; t - t')$ . Let us separate the total Hamiltonian  $\hat{H}^{\text{tot}}$  into purely elastic part  $\hat{H}^{\text{el}}$  and non-elastic part  $\hat{H}^{\text{non}}$ . We will discuss elastic Hamiltonian  $\hat{H}^{\text{el}}$  in details in the next section. Subtracting elastic Hamiltonian we define non-elastic stress tensor which comes from non-elastic Hamiltonian  $\hat{H}^{\text{non}}$ :

$$\begin{aligned} \hat{H}^{\text{non}} &= \hat{H}_0^{\text{non}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{non}}(\vec{x}) + \mathcal{O}(e_{ij}^2) \\ \hat{T}_{ij}^{\text{non}}(\vec{x}) &= \frac{\delta \hat{H}^{\text{non}}}{\delta e_{ij}(\vec{x})} \end{aligned} \quad (3.5)$$

In the rest of this thesis we will always use  $\hat{H}_0$ ,  $\chi_{ijkl}$  and  $\hat{T}_{ij}$  to represent non-elastic quantities  $\hat{H}_0^{\text{non}}$ ,  $\chi_{ijkl}^{\text{non}}$  and  $\hat{T}_{ij}^{\text{non}}$ , while use  $\hat{H}^{\text{el}}$ ,  $\chi_{ijkl}^{\text{el}}$  and  $\hat{T}_{ij}^{\text{el}}$  to represent the elastic Hamiltonian, susceptibility and stress tensor. Define eigenbasis of non-elastic Hamiltonian  $\hat{H}_0$  to be  $|m\rangle$ , which is a generic multiple-level-system. We apply linear response theory to calculate space-averaged non-elastic susceptibility  $\chi_{ijkl}(\omega) = \frac{1}{L^3} \int d^3x d^3x' \chi_{ijkl}(\vec{x} - \vec{x}'; \omega)$ . This space-averaged susceptibility is volume independent. We use the same language as tunneling-two-level-system, that the susceptibility can be expressed in relaxation and resonance susceptibilities. The relaxation susceptibility comes from the energy eigenvalue shift due to the diagonal matrix elements of time-dependent perturbation, while the resonance susceptibility comes from the transitions between different eigenstates due

to the off-diagonal matrix elements of perturbation Hamiltonian:

$$\begin{aligned}
\chi_{ijkl}(\omega) &= \frac{1}{1 - i\omega\tau} \chi_{ijkl}^{\text{rel}} + \chi_{ijkl}^{\text{res}}(\omega + i\eta) \\
\chi_{ijkl}^{\text{rel}} &= \frac{\beta}{L^3} \sum_n \int d^3x d^3x' \left( \sum_m P_n P_m \langle n | \hat{T}_{ij}(\vec{x}) | n \rangle \langle m | \hat{T}_{kl}(\vec{x}') | m \rangle - P_n \langle n | \hat{T}_{ij}(\vec{x}) | n \rangle \langle n | \hat{T}_{kl}(\vec{x}') | n \rangle \right) \\
\chi_{ijkl}^{\text{res}}(\omega + i\eta) &= \frac{1}{L^3 \hbar} \sum_n \sum_{l \neq n} \int d^3x d^3x' P_n \frac{\langle n | \hat{T}_{ij}(\vec{x}) | l \rangle \langle l | \hat{T}_{kl}(\vec{x}') | n \rangle}{\omega + (E_n - E_l)/\hbar + i\eta} \\
&\quad - \frac{1}{L^3 \hbar} \sum_l \sum_{n \neq l} \int d^3x d^3x' P_l \frac{\langle n | \hat{T}_{ij}(\vec{x}) | l \rangle \langle l | \hat{T}_{kl}(\vec{x}') | n \rangle}{\omega + (E_n - E_l)/\hbar + i\eta}
\end{aligned} \tag{3.6}$$

Where we use  $\frac{1}{1 - i\omega\tau} \chi_{ijkl}^{\text{rel}}$  and  $\chi_{ijkl}^{\text{res}}(\omega + i\eta)$  for relaxation and resonance susceptibilities.  $L^3$  is the volume,  $\omega$  is external strain field frequency,  $\tau$  is the effective thermal relaxation time for glass multiple-level-system  $\hat{H}_0$ ,  $E_n$  is the  $n$ -th eigenvalue and  $P_n = e^{-\beta E_n} / \mathcal{Z}$  the distribution probability of it. The non-elastic susceptibility obeys the generic form of arbitrary isotropic materials:  $\chi_{ijkl} = (\chi_l - 2\chi_t) \delta_{ij} \delta_{kl} + \chi_t (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk})$  [25], where  $\chi_l$  is compression modulus and  $\chi_t$  is shear modulus. Please note that the  $n$ -th eigenstate thermal relaxation process is the summation of all relaxation processes between different  $m$ -th levels and  $n$ -th level. The effective thermal relaxation time  $\tau_n$  should differ for various quantum numbers  $n = 0, 1, 2, \dots$ . However, in this thesis we focus on the real part of susceptibility, and we consider it in relaxation and resonance regimes separately. In relaxation regime with  $\omega\tau_n \ll 1$ , the factor  $(1 - i\omega\tau_n)^{-1}$  makes the imaginary part of relaxation susceptibility much smaller than real part of it, while in resonance regime with  $\omega\tau_n \gg 1$ ,  $(1 - i\omega\tau_n)^{-1}$  makes relaxation susceptibility negligible compared to the resonance susceptibility. The only regime sensitive to  $\tau_n$  is relaxation-resonance cross-over regime with  $\omega\tau_n \approx 1$ . Therefore in the relaxation susceptibility of Eq.(3.6) we use the approximation to replace  $\tau_n, \forall n = 0, 1, 2, \dots$  with multiple-level-system effective thermal relaxation time  $\tau$ .

## 3.2 Elastic Susceptibility

The elastic Hamiltonian  $\hat{H}^{\text{el}}$  is usually written in terms of phonon wave functions. It can be represented by phonon creation-annihilation operators:

$$\hat{H}^{\text{el}} = \sum_{k\alpha} \hbar\omega_{k\alpha} \left( \hat{a}_{k\alpha}^\dagger \hat{a}_{k\alpha} + \frac{1}{2} \right) \tag{3.7}$$

where  $\alpha$  is phonon polarization, i.e., longitudinal and transverse  $\alpha = l, t$ . Due to the definition of elastic stress tensor  $\hat{T}_{ij}^{\text{el}}(\vec{x}) = \delta \hat{H}^{\text{el}} / \delta e_{ij}(\vec{x})$ , the inverse of elastic stress-stress susceptibility is  $((\chi^{\text{el}})^{-1})_{ijkl}(x, x'; t, t') =$



$\delta e_{kl}(\vec{x}', t') / \delta \langle \hat{T}_{ij}^{\text{el}} \rangle(\vec{x}, t) = -\frac{i}{\hbar} \Theta(t-t') \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \langle m | [e_{ij}(\vec{x}, t), e_{kl}(\vec{x}', t')] | m \rangle$ , where  $\Theta(t-t')$  is time-ordered operator, and  $\mathcal{Z} = \sum_m e^{-\beta E_m}$  is partition function of phonon energy levels. The full elastic susceptibility containing higher order corrections from non-elastic susceptibility can be derived by Dyson equation:

$$\left[ (\chi^{\text{el}})^{-1} \right]_{ijkl}^{-1} \Big|_{\text{full}} (k; \omega) = \left[ (\chi^{\text{el}})^{-1} \right]_{ijkl}^{-1} (k; \omega) - \chi_{ijkl}(\omega) \quad (3.8)$$

where the inversed bare elastic susceptibility reads  $(\chi^{\text{el}})_{l,t}^{-1} = \frac{1}{\rho c_{l,t}^2} \frac{\omega_k^2}{\omega^2 - \omega_k^2}$ . The wave number independence of non-elastic susceptibility comes from the assumption we make later, that the non-elastic susceptibility is diagonal in spacial coordinates. Please see section 3.2.1 for details. From Eq.(3.8) we find the phonon frequency is shifted away from  $\omega_k = c_{l,t} k$ :

$$\frac{\Delta \omega_k}{\omega_k} = \frac{\text{Re } \chi_{l,t}(\omega) + i \text{Im } \chi_{l,t}(\omega)}{2\rho c_{l,t}^2} \quad (3.9)$$

where the real part of frequency shift corresponds to sound velocity shift, while the imaginary part is relate to internal friction  $Q^{-1}$ . From Eq.(3.6) non-elastic susceptibility has two parts, relaxation and resonance susceptibilities. In low temperature high frequency resonance regime,  $\omega\tau \gg 1$  so the prefactor of relaxation susceptibility  $(1 - i\omega\tau)^{-1}$  makes it negligible compared to the resonance one. The sound velocity shift is dominated by resonance susceptibility. In high temperature low frequency relaxation regime  $\omega\tau \ll 1$ , so  $(1 - i\omega\tau)^{-1} \approx 1$ , relaxation susceptibility is no longer much smaller than the resonance one. Both of relaxation and resonance susceptibilities contribute to relaxation regime sound velocity shift:

$$\begin{aligned} \frac{\Delta \omega_{k;l,t}}{\omega_{k;l,t}} &= \frac{\text{Re} \left( \chi_{l,t}^{\text{res}}(\omega) + \chi_{l,t}^{\text{rel}}(\omega) \right)}{2\rho c_{l,t}^2} && \text{relaxation regime} \\ \frac{\Delta \omega_{k;l,t}}{\omega_{k;l,t}} &= \frac{\text{Re } \chi_{l,t}^{\text{res}}(\omega)}{2\rho c_{l,t}^2} && \text{resonance regime} \end{aligned} \quad (3.10)$$

where  $\omega$  is corresponding phonon frequency. At the beginning of chapter 4, we will further discuss the elastic susceptibility.

### 3.2.1 Virtual Phonon Exchange Interactions

Within single-block considerations, non-elastic stress tensor  $\hat{T}_{ij}(\vec{x})$  is just a generalization of TLS model. However, if we combine a set of such blocks, the interaction between them will be taken into account. Since the stress-strain coupling  $e_{ij} \hat{T}_{ij}$  contains phonon strain  $e_{ij}$ , the exchange of virtual phonons will generate an

effective RKKY-type interaction between blocks via stress tensor products:

$$\hat{V} = \int d^3x d^3x' \sum_{ijkl} \Lambda_{ijkl}(\vec{x} - \vec{x}') \hat{T}_{ij}(\vec{x}) \hat{T}_{kl}(\vec{x}') \quad (3.11)$$

where the coefficient  $\Lambda_{ijkl}(\vec{x} - \vec{x}')$  was first derived by Joffrin and Levelut[42]. A further detailed correction to this coefficient was given by D. Zhou and A. J. Leggett[28]. Please see Appendix (A) for detailed derivations

$$\Lambda_{ijkl}(\vec{x} - \vec{x}') = -\frac{\tilde{\Lambda}_{ijkl}}{8\pi\rho c_t^2 |\vec{x} - \vec{x}'|^3} \quad (3.12)$$

$$\begin{aligned} \tilde{\Lambda}_{ijkl} = & \frac{1}{4} \left\{ (\delta_{jl} - 3n_j n_l) \delta_{ik} + (\delta_{jk} - 3n_j n_k) \delta_{il} + (\delta_{ik} - 3n_i n_k) \delta_{jl} + (\delta_{il} - 3n_i n_l) \delta_{jk} \right\} \\ & + \frac{1}{2} \left( 1 - \frac{c_t^2}{c_l^2} \right) \left\{ -(\delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{jk} \delta_{il}) \right. \\ & \left. + 3(n_i n_j \delta_{kl} + n_i n_k \delta_{jl} + n_i n_l \delta_{jk} + n_j n_k \delta_{il} + n_j n_l \delta_{ik} + n_k n_l \delta_{ij}) - 15n_i n_j n_k n_l \right\} \end{aligned} \quad (3.13)$$

where  $\vec{n}$  is the unit vector of  $\vec{x} - \vec{x}'$ , and  $i, j, k, l$  runs over 1, 2, 3 cartesian coordinates. We call Eq.(3.11) non-elastic stress-stress interaction. In the rest of this thesis we always 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, when  $\vec{x}_s$  denotes the center of the  $s$ -th block, and that  $\int_{V^{(s)}} \hat{T}_{ij}(\vec{x}) d^3x = \hat{T}_{ij}^{(s)}$  is the uniform stress tensor of the  $s$ -th block. Also we use  $e_{ij}^{(s)}(t)$  to denote the phonon strain field  $e_{ij}(\vec{x}, t)$  located at the  $s$ -th block. By combining  $N_0 \times N_0 \times N_0$  identical  $L \times L \times L$  unit blocks to form a  $N_0 L \times N_0 L \times N_0 L$  super block, the non-elastic Hamiltonian without external strain field is written as

$$\hat{H}^{\text{super}} = \sum_{s=1}^{N_0^3} \hat{H}_0^{(s)} + \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \quad (3.14)$$

From now on we make the assumption that block uniform stress tensors  $\hat{T}_{ij}^{(s)}$  correlation function (i.e., non-elastic susceptibility) is diagonal in spacial coordinates:  $\chi_{ijkl}^{(ss')} = L^{-3} \langle \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \rangle = \chi_{ijkl} \delta_{ss'}$ .

### 3.3 Glass Full Hamiltonian with the Presence of External Strain

In this section we consider glass super block Hamiltonian with the presence of external strain field  $\mathbf{e}(\vec{x}, t)$  as a perturbation. Please note that we have defined non-elastic stress tensor and non-elastic stress-stress susceptibility with the help of intrinsic phonon strain field, in this section  $\mathbf{e}(\vec{x}, t)$  stands for the external real

phonon field. It seems the Hamiltonian Eq.(3.14) simply adds a stress-strain coupling  $\sum_{s=1}^{N_0^3} \sum_j e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)}$ . However, more questions arise with the presence of  $\mathbf{e}(\vec{x}, t)$ . First of all, in Eq.(3.5) we expand non-elastic Hamiltonian  $\hat{H}^{(s)}$  in orders of strain field. The zeroth order term  $\hat{H}_0^{(s)}$  is by definition not the function of  $\mathbf{e}(\vec{x}, t)$ , which means the eigenstates  $|n^{(s)}\rangle$  and eigenvalues  $E_n^{(s)}$  of  $\hat{H}_0^{(s)}$  are unaffected by external field.

Second,  $\hat{T}_{ij}^{(s)}$  might be modified by  $\mathbf{e}(\vec{x}, t)$ . A familiar example is that external strain field can modify electric dipole moments by changing positive-negative charge pairs' relative positions:  $\Delta p_i = \sum_j (\partial u_i / \partial x_j) p_j$ . Let's denote the change of  $\hat{T}_{ij}^{(s)}$  is  $\Delta \hat{T}_{ij}^{(s)}(\mathbf{e})$ . We further define new stress tensor operator  $\hat{T}_{ij}^{(s)}(\mathbf{e})$  as follows, as the functional derivative of non-elastic Hamiltonian at the presence of external real strain with respect to intrinsic strain field  $e_{ij}^{(s)}$ :

$$\hat{T}_{ij}^{(s)}(\mathbf{e}) = \hat{T}_{ij}^{(s)} + \Delta \hat{T}_{ij}^{(s)}(\mathbf{e}) = \delta \hat{H}^{(s)}(\mathbf{e}) / \delta e_{ij}^{(s)} \quad (3.15)$$

which means the new operator  $\hat{T}_{ij}^{(s)}(\mathbf{e})$  is non-elastic stress tensor under the presence of external strain field  $\mathbf{e}$ . The stress-strain coupling is then given by  $\sum_s \sum_{ij} e_{ij}^{(s)} \hat{T}_{ij}^{(s)}(\mathbf{e})$ , where  $\mathbf{e}$  is external phonon strain field. The non-elastic susceptibility is given by replacing  $\hat{T}_{ij}^{(s)}$  with  $\hat{T}_{ij}^{(s)}(\mathbf{e})$ . The exchange of virtual phonon gives non-elastic stress-stress interaction  $\hat{V} = \sum_{ss'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)}(\mathbf{e}) \hat{T}_{kl}^{(s')}(\mathbf{e})$ . In the rest of this chapter, we write  $\hat{T}_{ij}^{(s)}$  to stand for  $\hat{T}_{ij}^{(s)}(\mathbf{e})$  for simplicity, where  $\mathbf{e}$  is not virtual phonon strain field, but external real strain field.

Finally the relative positions of blocks  $\vec{x}^{(s)} - \vec{x}^{(s')}$  are changed by external strain field, so that the coefficient of non-elastic stress-stress interaction is modified from  $\Lambda_{ijkl}^{(ss')}$  to  $\Lambda_{ijkl}^{(ss')}(\mathbf{e})$ . Thus the glass non-elastic Hamiltonian is  $\hat{H}^{\text{super}}(\mathbf{e}) = \sum_s \left( \hat{H}_0^{(s)} + \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)} \right) + \sum_{s \neq s'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')}(\mathbf{e}) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}$ . The super block non-elastic stress tensor is given by  $\hat{T}_{ij}^{\text{super}} = \delta \hat{H}^{\text{super}}(\mathbf{e}) / \delta e_{ij}$ . Because  $\Lambda_{ijkl}^{(ss')}(\mathbf{e})$  is the function of external strain field, an extra term appears in super block stress tensor:

$$\hat{T}_{ij}^{\text{super}} = \sum_s e^{i\vec{k} \cdot \vec{x}_s} \hat{T}_{ij}^{(s)} + \sum_{s \neq s'} \sum_{abcd} e^{i\vec{k} \cdot \frac{\vec{x}_s + \vec{x}_{s'}}{2}} \frac{\delta \Lambda_{abcd}^{(ss')}(\mathbf{e})}{\delta e_{ij}} \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} \quad (3.16)$$

where the above result is obtained in long wavelength limit. We therefore rewrite super block Hamiltonian as the summation of unperturbed part  $\hat{H}_0^{\text{super}}(\mathbf{e})$  and time-dependent perturbation  $\sum_{ij} e_{ij}(t) \hat{T}_{ij}^{\text{super}}$ :

$$\begin{aligned} \hat{H}^{\text{super}}(\mathbf{e}) &= \hat{H}_0^{\text{super}}(\mathbf{e}) + \sum_{ij} e_{ij}(t) \hat{T}_{ij}^{\text{super}} \\ \hat{H}_0^{\text{super}}(\mathbf{e}) &= \sum_{s=1}^{N_0^3} \hat{H}_0^{(s)} + \sum_{s \neq s'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} (0) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \end{aligned} \quad (3.17)$$

Please note, that in Eq.(3.17)  $e(t)$  is real phonon strain field. Similar with super block non-elastic stress tensor, the non-elastic susceptibility  $\chi_{ijkl}^{\text{super}}(\omega)$  receives an extra term as well. To calculate super block non-elastic susceptibility let us denote  $|n^*\rangle$  and  $E_n^*$  to be the  $n$ -th eigenstate and eigenvalue for super block unperturbed Hamiltonian  $\hat{H}_0^{\text{super}}(e)$ , and use linear response theory with respect to perturbation  $\sum_{ij} e_{ij}(t)\hat{T}_{ij}^{\text{super}}$ . Please note when deriving super block relaxation susceptibility, the ‘‘effective thermal relaxation time’’  $\tau^{\text{super}}$  should be different from that of unit blocks  $\tau$ . However, since we will be only interested in relaxation regime with  $\omega\tau, \omega\tau^{\text{super}} \ll 1$  and resonance regime with  $\omega\tau, \omega\tau^{\text{super}} \gg 1$  separately, the exact relation between  $\tau$  and  $\tau^{\text{super}}$  is not important. We still use  $\tau$  to stand for super block relaxation time for convenience. The super block non-elastic susceptibility is given by

$$\begin{aligned} \chi_{ijkl}^{\text{super}}(\omega) &= \frac{1}{(N_0L)^3} \frac{\beta}{1-i\omega\tau} \left( \sum_{n^*m^*} \frac{e^{-\beta(E_n^*+E_m^*)}}{\mathcal{Z}^{*2}} \langle n^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle m^* | \hat{T}_{kl}^{\text{super}} | m^* \rangle \right. \\ &\quad \left. - \sum_{n^*} \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \langle n^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle n^* | \hat{T}_{kl}^{\text{super}} | n^* \rangle \right) \\ &\quad + \frac{1}{(N_0L)^3} \frac{2}{\hbar} \sum_{n^*l^*} \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \frac{(E_l^* - E_n^*)/\hbar}{(\omega + i\eta)^2 - (E_l^* - E_n^*)^2/\hbar^2} \langle l^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle n^* | \hat{T}_{kl}^{\text{super}} | l^* \rangle \quad (3.18) \end{aligned}$$

where  $\hat{T}_{ij,cc}^{\text{super}}$  is the complex conjugate of  $\hat{T}_{ij}^{\text{super}}$ . The first line of Eq.(3.18) is super block non-elastic relaxation susceptibility  $\frac{1}{1-i\omega\tau}\chi_{ijkl}^{\text{super rel}}$ , and the second line is resonance susceptibility  $\chi_{ijkl}^{\text{super res}}(\omega + i\eta)$ .  $\mathcal{Z}^* = \sum_{n^*} e^{-\beta E_n^*}$  is distribution function of super block unperturbed Hamiltonian  $\hat{H}_0^{\text{super}}(e)$ .

## Chapter 4

# Universal Shift of Sound Velocity and Dielectric Constant in Glass

Before discussing the problems of low-temperature glass, let us first give a detailed discussion about the glass mechanical susceptibility. First of all, let us write the Hamiltonian of low-temperature glass as  $\hat{H}^{\text{tot}}$ . So the question is, what is contained in  $\hat{H}^{\text{tot}}$ ? First of all, long wavelength phonon Hamiltonian must be contained in it. The long wavelength phonon Hamiltonian can be represented by phonon creation/annihilation operators, given as follows,

$$\sum_{k\alpha} \hbar\omega_{k\alpha} \left( \hat{a}_{k\alpha}^\dagger \hat{a}_{k\alpha} + \frac{1}{2} \right) \quad (4.1)$$

where  $\alpha = l, t$  denotes the longitudinal and transverse phonon modes. Let us denote the above Hamiltonian Eq.(4.1) as the “purely elastic part of glass Hamiltonian”,  $\hat{H}^{\text{el}}$ . Subtracting the purely elastic part of Hamiltonian, the left-over glass Hamiltonian,  $\hat{H}^{\text{tot}} - \hat{H}^{\text{el}}$ , we call it “the non-elastic part of glass Hamiltonian”. We denote the non-elastic part Hamiltonian  $\hat{H}^{\text{tot}} - \hat{H}^{\text{el}}$  as  $\hat{H}^{\text{non}}$ .

According to D. C. Vural and A. J. Leggett[15], next we define the “strain operator”  $e_{ij}$  as follows: let us consider a cube of an arbitrary isotropic amorphous material, with the dimension  $L$  which is assumed large compared to “microscopic” lengths  $a$ , such as the typical interatomic distance, but is otherwise arbitrary. We define for such a block the strain operator  $e_{ij}$  in the standard way: if  $\vec{u}(\vec{x})$  denotes the displacement relative to some arbitrary reference frame of the matter at point  $\vec{x}$ , then

$$e_{ij}(\vec{x}) = \frac{1}{2} \left( \frac{\partial u_i(\vec{x})}{\partial x_j} + \frac{\partial u_j(\vec{x})}{\partial x_i} \right) \quad (4.2)$$

In the above discussions, we have defined the glass Hamiltonian  $\hat{H}^{\text{tot}}$ , purely elastic Hamiltonian  $\hat{H}^{\text{el}}$ , non-elastic Hamiltonian  $\hat{H}^{\text{non}}$  and strain  $e_{ij}(\vec{x})$ . Our next step is to expand the glass Hamiltonian,  $\hat{H}^{\text{tot}}$ , in

a Taylor series in the strain  $e_{ij}$ :

$$\hat{H}^{\text{tot}} = \hat{H}_0^{\text{tot}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{tot}}(\vec{x}) + \mathcal{O}(e^2) \quad (4.3)$$

In the above expansion, we define the following quantities:  $\hat{H}_0^{\text{tot}}$  is the leading order of glass total Hamiltonian in the Taylor series of strain;  $\int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{tot}}(\vec{x})$  is the first order of glass total Hamiltonian in the Taylor series of strain; the operator  $\hat{T}_{ij}^{\text{tot}}(\vec{x})$  is defined by  $\hat{T}_{ij}^{\text{tot}}(\vec{x}) = \delta \hat{H}^{\text{tot}} / \delta e_{ij}(\vec{x})$  as the coefficient of the first order expansion of glass total Hamiltonian in Taylor series. Throughout this thesis, we call this quantity “the glass total stress tensor operator”.

Next, let us expand the glass purely elastic Hamiltonian  $\hat{H}^{\text{el}}$  and non-elastic Hamiltonian  $\hat{H}^{\text{non}} = \hat{H}^{\text{tot}} - \hat{H}^{\text{el}}$  in the Taylor series in the strain  $e_{ij}$ :

$$\begin{aligned} \hat{H}^{\text{el}} &= \hat{H}_0^{\text{el}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{el}}(\vec{x}) + \mathcal{O}(e^2) \\ \hat{H}^{\text{non}} &= \hat{H}_0^{\text{non}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{non}}(\vec{x}) + \mathcal{O}(e^2) \end{aligned} \quad (4.4)$$

In the above expansion, we define the following quantities:  $\hat{H}_0^{\text{el}}$  and  $\hat{H}_0^{\text{non}}$  are the leading order of glass purely elastic and non-elastic Hamiltonians in the Taylor series of strain, respectively;  $\int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{el}}(\vec{x})$  and  $\int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{non}}(\vec{x})$  are the first order of glass purely elastic and non-elastic Hamiltonians in the Taylor series of strain, respectively; the operators  $\hat{T}_{ij}^{\text{el}}(\vec{x})$  and  $\hat{T}_{ij}^{\text{non}}(\vec{x})$  are defined by  $\hat{T}_{ij}^{\text{el}}(\vec{x}) = \delta \hat{H}^{\text{el}} / \delta e_{ij}(\vec{x})$  and  $\hat{T}_{ij}^{\text{non}}(\vec{x}) = \delta \hat{H}^{\text{non}} / \delta e_{ij}(\vec{x})$ . They are the coefficients of the first order expansion of glass purely elastic and non-elastic Hamiltonians in Taylor series, respectively. Throughout this thesis, we call these quantities “the glass purely elastic stress tensor  $\hat{T}_{ij}^{\text{el}}$ ” and “the glass non-elastic stress tensor  $\hat{T}_{ij}^{\text{non}}$ ”, respectively. According to the above definitions, the stress tensor operators have the simple relation:  $\hat{T}_{ij}^{\text{tot}}(\vec{x}) = \hat{T}_{ij}^{\text{el}}(\vec{x}) + \hat{T}_{ij}^{\text{non}}(\vec{x})$ .

Next, let us consider an externally imposed infinitesimal sinusoidal strain field,

$$e_{ij}(\vec{x}, t) = e_{ij} \left( e^{i\vec{k}\cdot\vec{x} - i\omega t} + e^{-i\vec{k}\cdot\vec{x} + i\omega t} \right) \quad (4.5)$$

where  $e_{ij}$  is real. The glass Hamiltonian  $\hat{H}^{\text{tot}}$  will provide a corresponding stress response of the material. We denote the response as  $\langle \hat{T}_{ij}^{\text{tot}} \rangle(\vec{x}, t)$ .

$$\langle \hat{T}_{ij}^{\text{tot}} \rangle(\vec{x}, t) = \langle \hat{T}_{ij}^{\text{tot}} \rangle e^{i\vec{k}\cdot\vec{x} - i\omega t} + c.c \quad (4.6)$$

where  $\langle \hat{T}_{ij}^{\text{tot}} \rangle$  is in general complex. Then we can define the complex response function  $\chi_{ij,kl}^{\text{tot}}(\vec{k}, \omega)$  in the standard way[2]

$$\chi_{ij,kl}^{\text{tot}}(\vec{k}, \omega) = \frac{\delta \langle \hat{T}_{ij}^{\text{tot}} \rangle}{\delta e_{kl}}(\vec{k}, \omega) = \frac{\delta^2 \langle \hat{H}^{\text{tot}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{tot}}(\vec{x}) \rangle}{\delta e_{ij} \delta e_{kl}}(\vec{k}, \omega) \quad (4.7)$$

Please note, that in the above definition of glass total susceptibility, Eq.(4.7), the glass overall Hamiltonian's expectation value (i.e., the Hamiltonian of glass which takes everything into account, including the glass total Hamiltonian, and the externally applied time-dependent perturbation) is defined by the glass total Hamiltonian  $\hat{H}^{\text{tot}}$  plus the time-dependent perturbation  $\int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{tot}}(\vec{x})$ ; the glass total stress response  $\langle \hat{T}_{ij}^{\text{tot}}(\vec{x}) \rangle(\vec{x}, t)$  is also defined by using the glass overall Hamiltonian: the summation of glass total Hamiltonian  $\hat{H}^{\text{tot}}$  and the perturbation:  $\hat{H}^{\text{tot}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{tot}}(\vec{x})$ .

On the other hand, the stress tensor have the relation:  $\hat{T}_{ij}^{\text{tot}}(\vec{x}) = \hat{T}_{ij}^{\text{el}}(\vec{x}) + \hat{T}_{ij}^{\text{non}}(\vec{x})$ . We can separate the glass total stress tensor into the purely elastic part and non-elastic part. The complex response function is therefore given by

$$\begin{aligned} \chi_{ij,kl}^{\text{tot}}(\vec{k}, \omega) &= \frac{\delta \langle \hat{T}_{ij}^{\text{tot}} \rangle}{\delta e_{kl}}(\vec{k}, \omega) = \frac{\delta \left( \langle \hat{T}_{ij}^{\text{el}} \rangle(\vec{k}, \omega) \right) + \delta \left( \langle \hat{T}_{ij}^{\text{non}} \rangle(\vec{k}, \omega) \right)}{\delta e_{kl}} \\ &= \frac{\delta^2 \langle \hat{H}^{\text{el}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{el}}(\vec{x}) \rangle}{\delta e_{ij} \delta e_{kl}}(\vec{k}, \omega) + \frac{\delta^2 \langle \hat{H}^{\text{non}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{non}}(\vec{x}) \rangle}{\delta e_{ij} \delta e_{kl}}(\vec{k}, \omega) \\ &= \chi_{ij,kl}^{\text{el}}(\vec{k}, \omega) + \chi_{ij,kl}^{\text{non}}(\vec{k}, \omega) \end{aligned} \quad (4.8)$$

In the rest of this thesis, we name  $\chi_{ij,kl}^{\text{el}}(\vec{k}, \omega)$  the purely elastic part of glass susceptibility; we name  $\chi_{ij,kl}^{\text{non}}(\vec{k}, \omega)$  the non-elastic part of glass susceptibility. Please note, that in the above definitions of glass elastic/non-elastic susceptibilities, Eq.(4.8), the glass elastic/non-elastic Hamiltonians' expectation values are defined by the glass elastic/non-elastic Hamiltonians  $\hat{H}^{\text{el}}$  and  $\hat{H}^{\text{non}}$  plus the time-dependent perturbation  $\int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{el}}(\vec{x})$  and  $\int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{non}}(\vec{x})$ ; the glass elastic/non-elastic stress responses  $\langle \hat{T}_{ij}^{\text{el}}(\vec{x}) \rangle(\vec{x}, t)$  and  $\langle \hat{T}_{ij}^{\text{non}}(\vec{x}) \rangle(\vec{x}, t)$  are also defined by using the glass elastic/non-elastic Hamiltonians plus the external time-dependent perturbations:  $\hat{H}^{\text{el}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{el}}(\vec{x})$  and  $\hat{H}^{\text{non}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{non}}(\vec{x})$ .

From the above definitions of elastic and non-elastic glass susceptibilities, at the static limit they are by definition negative. According to the definition in Eq.(4.8), the elastic susceptibility at static limit is given by

$$\chi_{ij,kl}^{\text{el}} = -(\rho c_t^2 - 2\rho c_l^2) \delta_{ij} \delta_{kl} - \rho c_t^2 (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) \quad (4.9)$$

The above result seems to be negative compared to the “elastic constant” in a standard elasticity textbook:  $(\rho c_l^2 - 2\rho c_t^2) \delta_{ij} \delta_{kl} + \rho c_t^2 (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk})$ . This is because: in the standard elasticity textbook, one usually defines the “elastic constant” through the definition  $\chi_{ijkl}^{\text{elastic constant}} = \delta^2 \langle \hat{H}^{\text{el}} \rangle / \delta e_{ij} \delta e_{kl}$ , but the Hamiltonian  $\hat{H}^{\text{el}}$  here is the elastic part of glass Hamiltonian which does not include the time-dependent perturbation  $\int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{el}}(\vec{x})$ . Therefore our definition of elastic susceptibility differs by a negative sign compared to the standard elastic constant in the standard textbook.

The non-elastic susceptibility, given by the definition in Eq.(4.8), is also negative at the static limit:

$$\begin{aligned}
\chi_{ijkl}^{\text{non}}(\omega) &= \frac{1}{1 - i\omega\tau} \chi_{ijkl}^{\text{non rel}} + \chi_{ijkl}^{\text{non res}}(\omega + i\eta) \\
\chi_{ijkl}^{\text{non rel}} &= \frac{\beta}{L^3} \sum_n \int d^3x d^3x' \left( \sum_m P_n P_m \langle n | \hat{T}_{ij}^{\text{non}}(\vec{x}) | n \rangle \langle m | \hat{T}_{kl}^{\text{non}}(\vec{x}') | m \rangle - P_n \langle n | \hat{T}_{ij}^{\text{non}}(\vec{x}) | n \rangle \langle n | \hat{T}_{kl}^{\text{non}}(\vec{x}') | n \rangle \right) \\
\chi_{ijkl}^{\text{non res}}(\omega + i\eta) &= \frac{1}{L^3 \hbar} \sum_n \sum_{l \neq n} \int d^3x d^3x' P_n \frac{\langle n | \hat{T}_{ij}^{\text{non}}(\vec{x}) | l \rangle \langle l | \hat{T}_{kl}^{\text{non}}(\vec{x}') | n \rangle}{\omega + (E_n - E_l)/\hbar + i\eta} \\
&\quad - \frac{1}{L^3 \hbar} \sum_l \sum_{n \neq l} \int d^3x d^3x' P_l \frac{\langle n | \hat{T}_{ij}^{\text{non}}(\vec{x}) | l \rangle \langle l | \hat{T}_{kl}^{\text{non}}(\vec{x}') | n \rangle}{\omega + (E_n - E_l)/\hbar + i\eta}
\end{aligned} \tag{4.10}$$

where  $|m\rangle$  and  $E_m$  are the  $m$ -th eigenstate and eigenvalue of the non-elastic part of glass Hamiltonian,  $\hat{H}^{\text{non}}$ .  $P_m = e^{-\beta E_m} / \mathcal{Z}$  is the  $m$ -th level probability function.  $\mathcal{Z}$  is the partition function of the non-elastic part of glass Hamiltonian.

One may ask the question, that in the most textbooks of elasticity theory, it seems more natural to define the “external stress” as the external field we apply on a certain material rather than the external strain  $e_{ij}(\vec{x}, t)$ . Suppose we apply an external stress on the material. To balance with the external stress, the material must provide an internal stress  $\langle \hat{T}_{ij}^{\text{tot}} \rangle(\vec{x}, t)$  by deforming itself. It will give rise to a corresponding strain response  $e_{ij}(\vec{x}, t)$ . We can also define the complex response function  $\chi_{ij,kl}^{\text{tot}} = \frac{\partial \langle \hat{T}_{ij}^{\text{tot}} \rangle}{\partial e_{kl}}(\vec{k}, \omega)$  by applying external stress as the external field. At the first glance the previous definition of response function seems to be different from the definition of response function by applying external strain as the external field. However, in the following discussions, we will see these two definitions are equivalent.

Suppose we apply an external stress on the material, the material must provide a deformation response  $e_{ij}(\vec{x}, t)$ , to generate an internal stress  $\langle \hat{T}_{ij}^{\text{tot}} \rangle(\vec{x}, t)$  which balances the external stress. On the contrary, instead of applying an external stress, we apply an external strain on the material. The material generates an internal stress  $\langle \hat{T}_{ij}^{\text{tot}} \rangle(\vec{x}, t)$ . To maintain the deformation, we must provide an external stress to balance the internal stress  $\langle \hat{T}_{ij}^{\text{tot}} \rangle(\vec{x}, t)$ . It is at this point that the two definitions of response function are equivalent. In the typical elasticity textbook we prefer to put in the external stress as the “external field”, then we measure



the material’s strain response; in this thesis, we prefer to put in the external strain as the “external field”, then we measure the material’s internal stress response. Both definitions give the same glass mechanical response function.

One of the unambiguous evidences presented by Zeller and Pohl[1] is that the low temperature heat capacity of glass differs significantly from that of crystalline solids. In pure and defect-free insulating crystals the heat capacity is proportional to  $T^3$  below 1K, which comes from phonon vibration modes. However in glass the heat capacity is the summation of two parts: long wavelength phonon contribution from Debye’s theory, and an excess specific heat known as the glass excitations approximated by  $C_{\text{excess}} = c_1 T^{1+\delta} + c_2 T^3$ , where  $\delta < 1$ ,  $c_1$  and  $c_2$  varies for different materials[2]. Anderson, Halperin and Varma[3] group and Phillips[19] independently developed a model which was later known as tunneling-two-level-system (TTLS) (see chapter 2). It successfully explained glass excess heat capacity, together with several other universal properties such as saturation, echoes etc.

To further verify the existence of two-level-systems, L. Piché, R. Maynard, S. Hunklinger and J. Jäckle[33] studied the influence of two-level-systems on the variation of the sound velocity of longitudinal waves in vitreous silica Suprasil I at temperatures  $0.28\text{K} < T < 4.2\text{K}$  and frequencies  $30\text{MHz} < f < 150\text{MHz}$ . The sound velocity shift was found to be logarithmically dependent on temperature. In the high frequency low temperature resonance regime with  $\omega\tau \gg 1$  ( $\tau$  is the effective thermal relaxation time, please refer to section 2(A) for detailed discussions) the sound velocity increases with increasing temperature. This sound velocity shift in resonance regime is independent of phonon frequency. In the low frequency high temperature relaxation regime with  $\omega\tau \ll 1$  the velocity decreases with increasing temperature. Such sound velocity increase-decrease transition occurs at the transition point  $\omega\tau(T) \approx 1$ , which means the transition temperature  $T$  is functional of phonon frequency. However, as long as the sound velocity measurement enters into relaxation regime, it turns out to be frequency independent as well. In the rest of this thesis we will discuss the slope of  $\ln T$  dependence of sound velocity in relaxation and resonance regimes separately, so we assume that sound velocity shift is frequency independent in both relaxation and resonance regimes. Such universality has been observed in amorphous materials such as vitreous silica, lithium-doped KCl[4] and silica based microscopic cover glass[8], etc..

By averaging over random parameters of glass two-level-system susceptibility, TTLS model successfully explained the logarithmic temperature dependence of sound velocity shift[19, 10] (see chapter 2). It also proves that the slope of  $\ln T$  dependence is negative in relaxation regime and positive in resonance regime. The sound velocity slope ratio between relaxation and resonance regimes is  $\mathcal{C}^{\text{rel}} : \mathcal{C}^{\text{res}} = -1/2 : 1$ , which agrees quite well with silica based microscopic cover glass measurements[8]. However at least to the author’s

knowledge, it's the only amorphous material with the absolute value of the slope in relaxation regime smaller than that of resonance regime: other materials, present the absolute value of the slope in relaxation regime equal or slightly greater than that of resonance regime: vitreous silica Suprasil I[33], PdSiCu[9], Zr-Nb[17], lithium-doped KCl[4], vitreous silica[11], metallic glass[13] Ni<sub>81</sub>P<sub>19</sub>, etc. (the electron-TTLS coupling in metallic glass is relatively weak compared to phonon-TTLS coupling, so conducting electrons are not strong enough to affect sound velocity[10]). S. Hunklinger and C. Enss[12] suggest that most of the sound velocity slope ratios of glass materials are rather  $-1$  to  $1$ , probably due to the interaction between tunneling systems, because glass defects are highly concentrated. In this chapter our main goal is to set up a generic coupled block model to discuss the universal property of temperature dependence on sound velocity shift.

By assuming that electric field couples to two-level-systems[7], the result of TTLS model on dielectric constant shift is similar with sound velocity shift, but the dielectric shift slope ratio between relaxation and resonance regimes is  $\mathcal{C}^{\text{rel}} : \mathcal{C}^{\text{res}} = +1/2 : -1$ . However, dielectric measurements on various amorphous materials such as vitreous silica Suprasil W and vitreous As<sub>2</sub>S<sub>3</sub>[14], vitreous silica Suprasil I[6] and borosilicate glass (BK7)[15] indicate that the slope ratio is  $\mathcal{C}^{\text{rel}} : \mathcal{C}^{\text{res}} = +1 : -1$ , regardless of their microscopic nature. At the end of this chapter we use electric dipole-dipole interaction to discuss such universal shift of glass dielectric constant.

In this chapter we want to focus on the universal shift of glass sound velocity and dielectric constant by developing a theory of coupled generic blocks. From chapter 2 we have set up our generic coupled block model, by expanding non-elastic part of glass Hamiltonian in orders of intrinsic strain field  $e_{ij}(\vec{x}, t)$  and putting in virtual phonon exchange interaction. In this chapter our goal is to set up the renormalization recursion relation between large and small length scale non-elastic susceptibilities. We want to prove for different kinds of amorphous materials, at experimental large length scale the sound velocity and dielectric constant shift in relaxation and resonance regimes have the same universal behavior, regardless of their microscopic properties. However, as we will see from the renormalization equations of relaxation and resonance susceptibilities, we are not able to prove universal shift of sound velocity and dielectric constant, mainly because the negativity of relaxation and resonance susceptibilities which leads to the increasing behavior of them as the length scale increases than the expected decreasing behavior of susceptibilities.

## 4.1 Renormalization of Glass Non-Elastic Susceptibility

We have given a detailed discussion on single and super block non-elastic susceptibilities (see Eq.(3.6) and Eq.(3.18)). For convenience we write them down again as follows,

$$\begin{aligned}
\chi_{ijkl}(\omega) &= \frac{1}{1 - i\omega\tau} \chi_{ijkl}^{\text{rel}} + \chi_{ijkl}^{\text{res}}(\omega + i\eta) \\
\chi_{ijkl}^{\text{rel}} &= \frac{\beta}{L^3} \sum_n \int d^3x d^3x' \left( \sum_m P_n P_m \langle n | \hat{T}_{ij}(\vec{x}) | n \rangle \langle m | \hat{T}_{kl}(\vec{x}') | m \rangle - P_n \langle n | \hat{T}_{ij}(\vec{x}) | n \rangle \langle n | \hat{T}_{kl}(\vec{x}') | n \rangle \right) \\
\chi_{ijkl}^{\text{res}}(\omega + i\eta) &= \frac{1}{L^3 \hbar} \sum_n \sum_{l \neq n} \int d^3x d^3x' P_n \frac{\langle n | \hat{T}_{ij}(\vec{x}) | l \rangle \langle l | \hat{T}_{kl}(\vec{x}') | n \rangle}{\omega + (E_n - E_l)/\hbar + i\eta} \\
&\quad - \frac{1}{L^3 \hbar} \sum_l \sum_{n \neq l} \int d^3x d^3x' P_l \frac{\langle n | \hat{T}_{ij}(\vec{x}) | l \rangle \langle l | \hat{T}_{kl}(\vec{x}') | n \rangle}{\omega + (E_n - E_l)/\hbar + i\eta} \tag{4.11}
\end{aligned}$$

$$\begin{aligned}
\chi_{ijkl}^{\text{super}}(\omega) &= \frac{1}{1 - i\omega\tau} \chi_{ijkl}^{\text{super rel}} + \chi_{ijkl}^{\text{super res}}(\omega + i\eta) \\
&= \frac{1}{(N_0 L)^3} \frac{\beta}{1 - i\omega\tau} \left( \sum_{n^* m^*} \frac{e^{-\beta(E_n^* + E_m^*)}}{\mathcal{Z}^{*2}} \langle n^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle m^* | \hat{T}_{kl}^{\text{super}} | m^* \rangle \right. \\
&\quad \left. - \sum_{n^*} \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \langle n^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle n^* | \hat{T}_{kl}^{\text{super}} | n^* \rangle \right) \\
&\quad + \frac{1}{(N_0 L)^3} \frac{2}{\hbar} \sum_{n^* l^*} \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \frac{(E_l^* - E_n^*)/\hbar}{(\omega + i\eta)^2 - (E_l^* - E_n^*)^2/\hbar^2} \langle l^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle n^* | \hat{T}_{kl}^{\text{super}} | l^* \rangle \tag{4.12}
\end{aligned}$$

where in the last section we define the  $n$ -th eigenstate and eigenvalue of super block to be  $|n^*\rangle$  and  $E_n^*$ .

We also use  $\chi_{ijkl}$ ,  $\chi_{ijkl}^{\text{res}}$ ,  $\chi_{ijkl}^{\text{rel}}$ ,  $\chi_{ijkl}^{\text{super}}$ ,  $\chi_{ijkl}^{\text{super res}}$ ,  $\chi_{ijkl}^{\text{super rel}}$ ,  $\hat{T}_{ij}$  and  $\hat{T}_{ij}^{\text{super}}$  to stand for  $\chi_{ijkl}^{\text{non}}$ ,  $\chi_{ijkl}^{\text{non res}}$ ,  $\chi_{ijkl}^{\text{non rel}}$ ,  $\chi_{ijkl}^{\text{super non}}$ ,  $\chi_{ijkl}^{\text{super non res}}$ ,  $\chi_{ijkl}^{\text{super non rel}}$ ,  $\hat{T}_{ij}^{\text{non}}$  and  $\hat{T}_{ij}^{\text{super non}}$ .

In this section our goal is to set up the relation between single block and super block non-elastic susceptibilities. Since the unit and super block susceptibilities' length scales differ by a factor of  $N_0$ , repeating this real space renormalization carries out experimental large length scale non-elastic susceptibility eventually. The starting microscopic length scale of renormalization is, for example,  $L_1 \sim 50\text{\AA}$  by D. C. Vural and A. J. Leggett[25]. Since the final result only logarithmically depends on this choice, it will not be sensitive. In the  $n$ -th step of renormalization, we combine  $N_0^3$  identical blocks with the dimension  $L_n \times L_n \times L_n$  to form a  $n$ -th step super block with the dimension  $N_0 L_n \times N_0 L_n \times N_0 L_n$ . In the next step the unit block length scale is  $L_{n+1} = N_0 L_n$ . We begin with such a group of non-interacting unit blocks with bare Hamiltonian  $\hat{H}_0 = \sum_{s=1}^{N_0^3} \hat{H}_0^{(s)}$ , eigenstates  $|n\rangle = \prod_{s=1}^{N_0^3} |n^{(s)}\rangle$  and eigenvalues  $E_n = \sum_{s=1}^{N_0^3} E_n^{(s)}$ . Please note  $E_n^{(s)}$  stands for the  $s$ -th unit block eigenvalue for the  $n^{(s)}$ -th eigenstate. We combine them to form a super block and turn

on non-elastic stress-stress interaction  $\hat{V} = \sum_{s \neq s'}^{N_0^3} \Lambda_{ijkl}^{(ss')} (0) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}$ . We assume non-elastic stress-stress interactions  $\hat{V}$  are relatively weak compared to the summation of unit block Hamiltonians  $\hat{H}_0 = \sum_{s=1}^{N_0^3} \hat{H}_0^{(s)}$ , so that the interactions can be treated as a perturbation. If the non-elastic susceptibility decreases logarithmically as the increase of length scale, then that means the non-elastic stress-stress interaction  $\hat{V}$  can be treated as a perturbation at the late stages. The assumption that  $\hat{V}$  can be treated as a perturbation is qualitatively correct. The  $n$ -th eigenstate and eigenvalue of super block are  $|n^*\rangle$  and  $E_n^*$ . Their relations with  $|n\rangle$  and  $E_n$  are given as follows

$$\begin{aligned} |n^*\rangle &= |n\rangle + \sum_{p \neq n} \frac{\langle p | \hat{V} | n \rangle}{E_n - E_p} |p\rangle + \mathcal{O}(V^2) \\ E_n^* &= E_n + \langle n | V | n \rangle + \sum_{p \neq n} \frac{|\langle p | \hat{V} | n \rangle|^2}{E_n - E_p} |p\rangle + \mathcal{O}(V^2) \end{aligned} \quad (4.13)$$

With the help of Eq.(4.13) one can rewrite super block non-elastic susceptibility in terms of unit block susceptibilities: we expand super-block relaxation and resonance susceptibilities up to the first order of interaction  $\hat{V}$ , and sum over eigenstates  $|n\rangle = \prod_s |n^{(s)}\rangle$ . These  $N_0^3$  non-interacting unit blocks' partition function is  $\mathcal{Z} = \prod_s \mathcal{Z}^{(s)}$ , and their  $n$ -th level probability function is  $P_n = \prod P_n^{(s)}$ . We apply the assumption that unit block stress tensors' matrix element products are diagonal in spacial coordinates, i.e., for different unit blocks ( $\vec{x}_s \neq \vec{x}'_s$ ) stress tensors, their matrix element products vanish. The combination of diagonal and off-diagonal stress tensor matrix elements can be exactly rewritten in terms of unit block relaxation and resonance susceptibilities. Finally we obtain the recursion relation between super block and unit block susceptibilities as follows, where both of super block and unit block susceptibilities are implicitly functional of temperature:

$$\begin{aligned} \chi_{ijkl}^{\text{super}}(\omega) &= \frac{1}{1 - i\omega\tau} \left\{ \chi_{ijkl}^{\text{rel}} - \frac{L_n^3}{N_0^3} \left[ - \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} (0) e^{-ik \cdot (x_s - x'_s)} \right] \left( \chi_{ijab}^{\text{rel}} \chi_{cdkl}^{\text{rel}} + 2\chi_{ijab}^{\text{rel}} \chi_{cdkl}^{\text{res}}(0) \right) \right\} \\ &+ \chi_{ijkl}^{\text{res}}(\omega + i\eta) - \frac{L_n^3}{N_0^3} \left[ - \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} (0) e^{-ik \cdot (x_s - x'_s)} \right] \chi_{ijab}^{\text{res}}(\omega + i\eta) \chi_{cdkl}^{\text{res}}(\omega + i\eta) \\ &+ \frac{\beta^{-1} L_n^3}{N_0^3 (1 - i\omega\tau)} \sum_{ss'} \sum_{abcdefgh} \frac{\delta \Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta \Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \left( \chi_{abef}^{\text{rel}(1)} \chi_{cdgh}^{\text{rel}(1)} - \chi_{abef}^{\text{rel}(2)} \chi_{cdgh}^{\text{rel}(2)} \right) \\ &- \frac{L_n^3}{N_0^3} \sum_{ss'} \sum_{abcdefgh} \frac{1}{\pi^2} \frac{\delta \Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta \Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \left\{ \int \frac{(1 - e^{-\beta\hbar(\omega_s + \omega'_s)})}{(1 - e^{-\beta\hbar\omega_s})(1 - e^{-\beta\hbar\omega'_s})} \frac{\text{Im} \chi_{abef}^{\text{res}}(\omega_s) \text{Im} \chi_{cdgh}^{\text{res}}(\omega'_s)}{\hbar\omega_s + \hbar\omega_{s'} - \hbar\omega} d(\hbar\omega_s) d(\hbar\omega'_s) \right. \\ &\quad \left. + i(1 - e^{-\beta\hbar\omega})\pi \int \frac{\text{Im} \chi_{abef}^{\text{res}}(\omega_s) \text{Im} \chi_{cdgh}^{\text{res}}(\omega - \omega_s)}{(1 - e^{-\beta\hbar\omega_s})(1 - e^{-\beta\hbar(\omega - \omega_s)})} d(\hbar\omega_s) \right\} \end{aligned} \quad (4.14)$$

For details of the calculations please see appendix (B).  $\chi_{ijkl}^{\text{rel}(1)}$ ,  $\chi_{ijkl}^{\text{rel}(2)}$  in the third line of Eq.(4.14) are the

first and second parts of relaxation susceptibility defined as follows:

$$\begin{aligned}
\chi_{ijkl}^{\text{rel}(1)} &= \frac{\beta}{L^3} \sum_{nm} P_n P_m \langle n | \hat{T}_{ij} | n \rangle \langle m | \hat{T}_{kl} | m \rangle \\
\chi_{ijkl}^{\text{rel}(2)} &= \frac{\beta}{L^3} \sum_n P_n \langle n | \hat{T}_{ij} | n \rangle \langle n | \hat{T}_{kl} | n \rangle \\
\frac{\chi_{ijkl}^{\text{rel}}}{1 - i\omega\tau} &= \frac{1}{1 - i\omega\tau} \left( \chi_{ijkl}^{\text{rel}(1)} - \chi_{ijkl}^{\text{rel}(2)} \right)
\end{aligned} \tag{4.15}$$

The super block susceptibility  $\chi_{ijkl}(\omega)$  is not functional of momentum  $\vec{k}$ , because we take long phonon wavelength limit  $\vec{k} \rightarrow 0$  in the coefficient  $\lim_{k \rightarrow 0} \sum_{ss'} \Lambda_{ijkl}^{(ss')}(0) e^{ik \cdot (x_s - x'_s)}$  of Eq.(4.14).

In Eq.(4.14), the third and fourth line terms' volume dependences are different from others'. This is because of the  $1/r^3$  behavior of  $\Lambda_{ijkl}^{(ss')}$ . We first investigate the volume dependence of the third line term, which is proportional to  $\beta^{-1}$ . Using the expression  $\chi_{ijkl}^{\text{rel}(1,2)} = (\chi_l^{\text{rel}(1,2)} - 2\chi_t^{\text{rel}(1,2)})\delta_{ij}\delta_{kl} + \chi_t^{\text{rel}(1,2)}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$  and summing over the indices, the third line can be simplified as  $\beta^{-1}C_{l,t}^{(1,2)}(\chi_{l,t}^{\text{rel}(1,2)})^2/(1-i\omega\tau)\rho^2c_{l,t}^4L_n^3$ , where  $C_{l,t}^{(1,2)}$  are dimensionless constants of order 1. If we require that there is a critical length scale  $L_c^{\text{rel}}$ , beyond which the third line of Eq.(4.14) is smaller than unit block relaxation susceptibility, the upper limit of  $L_c^{\text{rel}}$  is,

$$L_c^{\text{rel}} < \left( \frac{k_B T}{\rho c_{l,t}^2} \right)^{\frac{1}{3}} \tag{4.16}$$

we further let the temperature  $T$  to take an extremely high value,  $T = 10^4\text{K}$  (in fact the low-temperature glass ultrasonic sound velocity shift measurements are below 50K). The upper limit of  $L_c^{\text{rel}}$  is  $4.6\text{\AA}$  which is still smaller than  $50\text{\AA}$ , the effective starting length scale of our generic coupled block model.

On the other hand, to investigate the volume dependence of the fourth line term of Eq.(4.14), we use the assumption that the reduced imaginary resonance susceptibility  $\text{Im} \tilde{\chi}_{ijkl}^{\text{res}}(\omega) = \text{Im} \chi_{ijkl}^{\text{res}}(\omega)/(1 - e^{-\beta\hbar\omega})$  is approximately a constant up to the frequency of  $\omega_c$  and temperatures of the order 10K. Integrating over frequency variables  $\omega_s, \omega'_s$  the fourth line term gives  $-C_{l,t} [\hbar\omega_c \ln(\frac{\omega_c}{\omega}) - i\pi\hbar\omega] \left( \text{Im} \tilde{\chi}_{l,t}^{\text{res}} \right)^2 / \rho^2 c_{l,t}^4 L_n^3$ , where we obtain this result by using the expression  $\text{Im} \tilde{\chi}_{ijkl}^{\text{res}} = (\text{Im} \tilde{\chi}_l^{\text{res}} - 2\text{Im} \tilde{\chi}_t^{\text{res}}) \delta_{ij}\delta_{kl} + \text{Im} \tilde{\chi}_t^{\text{res}} (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$ .  $C_{l,t}$  is a positive constant of order 1, and  $\omega$  is input phonon frequency[33, 34] of order  $\omega \sim 1\text{MHz}$ . If we require that there is a critical length scale  $L_c^{\text{res}}$ , beyond which the fourth line term of Eq.(4.14) is smaller than unit block resonance susceptibility, we need to calculate the order of magnitude for  $L_c^{\text{res}}$ . The upper limit of  $L_c^{\text{res}}$  can be obtained by taking  $\omega_c$  to an extremely high value,  $\omega_c \sim 10^{15}\text{Hz}$  which corresponds to

$T \sim 10^4\text{K}$ :

$$L_c^{\text{res}} < \left( \frac{1}{\rho c_{l,t}^2} \frac{\hbar \omega_c}{\ln(\omega_c/\omega)} \right)^{\frac{1}{3}} \sim 1.7\text{\AA} < L_1 = 50\text{\AA} \quad (4.17)$$

which means the upper limit of  $L_c^{\text{res}}$  with super high cut-off frequency  $10^{15}\text{Hz}$  is also smaller than the starting effective length scale of our model. Thus throughout the entire renormalization procedure the third and fourth line terms in Eq.(4.14) are always negligible compared to other terms. This agrees with the conclusion by D. Zhou and A. J. Leggett[28] that the contribution of  $\Lambda_{ijkl}^{(ss')}(\mathbf{e})$  modification to resonant energy absorption is renormalization irrelevant. Dropping the third and fourth lines in Eq.(4.14) and taking the summations over indices  $abcd$ , the renormalization equations for non-elastic relaxation and resonance susceptibilities are reduced to

$$\begin{aligned} \chi_{ijkl}^{\text{super}}(\omega) &= \frac{1}{1 - i\omega\tau} \left\{ \chi_{ijkl}^{\text{rel}} - \frac{L_n^3}{N_0^3} \left[ - \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')}(0) e^{-ik \cdot (x_s - x'_s)} \right] (\chi_{ijab}^{\text{rel}} \chi_{cdkl}^{\text{rel}} + 2\chi_{ijab}^{\text{rel}} \chi_{cdkl}^{\text{res}}(0)) \right\} \\ &+ \chi_{ijkl}^{\text{res}}(\omega + i\eta) - \frac{L_n^3}{N_0^3} \left[ - \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')}(0) e^{-ik \cdot (x_s - x'_s)} \right] \chi_{ijab}^{\text{res}}(\omega + i\eta) \chi_{cdkl}^{\text{res}}(\omega + i\eta) \quad (4.18) \end{aligned}$$

We calculate the coefficient  $\frac{L_n^3}{N_0^3} \left[ - \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')}(0) e^{-ik \cdot (x_s - x'_s)} \right]$  which appears in Eq.(4.18), in Appendix F. We use the expression, that resonance and relaxation susceptibilities are written in the form of  $\chi_{ijkl}^{\text{res,rel}} = (\chi_l^{\text{res,rel}} - 2\chi_t^{\text{res,rel}})\delta_{ij}\delta_{kl} + \chi_t^{\text{res,rel}}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$ . The renormalization equations are further simplified as follows,

$$\begin{aligned} \chi_{t,l}^{\text{super rel}} &= \chi_{t,l}^{\text{rel}} - \frac{1}{\rho c_{t,l}^2} \left[ (\chi_{t,l}^{\text{rel}})^2 + 2\chi_{t,l}^{\text{rel}} \chi_{t,l}^{\text{res}}(0) \right] \\ \chi_{t,l}^{\text{super res}}(\omega + i\eta) &= \chi_{t,l}^{\text{res}}(\omega + i\eta) - \frac{1}{\rho c_{t,l}^2} \left[ \chi_{t,l}^{\text{res}}(\omega + i\eta) \right]^2 \quad (4.19) \end{aligned}$$

Eqs.(4.19) are the most important results of this thesis.

We now examine the implications of these renormalization equations. At the first glance, it seems that the non-elastic resonance susceptibility presents usual marginally renormalization irrelevant behavior with the increase of length scale: by repeating renormalization procedure for the modulus of resonance susceptibility from starting small length scale  $L_1$  to experimental length scale  $R$  we get logarithmic length scale dependence as follows

$$\frac{1}{\chi_{l,t}^{\text{res}}(\omega + i\eta, R)} = \frac{\ln(R/L_1)}{\rho c_{l,t}^2} + \frac{1}{\chi_{l,t}^{\text{res}}(\omega + i\eta, L_1)} \quad (4.20)$$

where the experimental length scale is given by input phonon wavelength  $R = 2\pi c_{l,t}/\omega$ . Except for the experiments by L. Piché and his collaborators[33] that the frequencies vary from  $f = 30 \sim 150$  MHz, and by G. Bellesa and his group[13] that  $f = 150$  MHz, most of the input frequencies are  $f = 1 \sim 20$  kHz[4, 8, 9, 11]. The factor  $\ln(R/L_1)$  is not sensitive to input phonon frequency: as  $f$  varies from 1 kHz to 100 MHz, it only changes from 20 to 10. Therefore in Eq.(4.20) we neglect the second term of r.h.s.. The modulus of experimental length scale resonance susceptibility is dominated by  $\rho c_{l,t}^2/\ln(R/L_1)$ . For the choice of  $f \sim 1$  kHz,  $R$  is of order  $\sim 10$  m, so  $L_1/R$  is of order  $\sim \times 10^{-10} \ll 1/\ln(R/L_1)$ .

However, if we stare at the definitions of relaxation and resonance susceptibilities in Eq.(4.11), we find that they are not positive, but negative quantities. First of all, the resonance susceptibility is negative. For example, let us choose  $\omega + i\eta = 0$ , and let  $(kl) = (ij)$  in resonance susceptibility, to consider  $\chi_{ijij}^{\text{res}}(\omega + i\eta = 0)$ ,

$$\chi_{ijkl}^{\text{res}}(\omega + i\eta = 0) = \frac{1}{L^3} \sum_{nl} \left( \frac{P_n - P_l}{E_n - E_l} \right) |\langle n | \hat{T}_{ij} | l \rangle|^2 < 0 \quad (4.21)$$

The resonance susceptibility  $\chi_{ijij}^{\text{res}}(\omega + i\eta = 0)$  is negative mainly because  $P_n < P_l$  for arbitrary pair of levels  $n, m$  with  $E_n > E_l$ . Because of the negativity of resonance susceptibility, the renormalization equation of resonance susceptibility is actually not marginally irrelevant, but renormalization relevant (see the second equation of Eqs.(4.19)).

On the other hand, the relaxation susceptibility  $\chi_{ijkl}^{\text{rel}}$  is negative as well. To prove this result let us define  $\sum_n P_n \langle n | \hat{T}_{ij} | n \rangle = \bar{T}_{ij}$  to be the ‘‘average value of stress tensor  $\hat{T}_{ij}$ ’’, and define  $\sum_n P_n \langle n | \hat{T}_{ij} | n \rangle^2 = (\bar{T}_{ij}^2)$  to be the ‘‘average value of the square of stress tensor  $\hat{T}_{ij}$ ’’. For simplicity we still let  $(kl) = (ij)$  to consider the relaxation susceptibility. The relaxation susceptibility  $\chi_{ijij}^{\text{rel}}$  is rewritten as follows,

$$\begin{aligned} \chi_{ijij}^{\text{rel}} &= \frac{\beta}{L^3} \sum_n \int d^3x d^3x' \left( \sum_m P_n P_m \langle n | \hat{T}_{ij}(\vec{x}) | n \rangle \langle m | \hat{T}_{ij}(\vec{x}') | m \rangle - P_n \langle n | \hat{T}_{ij}(\vec{x}) | n \rangle \langle n | \hat{T}_{ij}(\vec{x}') | n \rangle \right) \\ &= \frac{\beta}{L^3} \left( \sum_n \sum_m P_n P_m \langle n | \hat{T}_{ij} | n \rangle \langle m | \hat{T}_{ij} | m \rangle - \sum_n P_n \langle n | \hat{T}_{ij} | n \rangle \langle n | \hat{T}_{ij} | n \rangle \right) \\ &= \frac{\beta}{L^3} \left( (\bar{T}_{ij})^2 - (\bar{T}_{ij}^2) \right) < 0 \end{aligned} \quad (4.22)$$

Therefore, the relaxation susceptibility is always negative as well. If we look back to the first renormalization equation of Eqs.(4.19), the linear term  $-\frac{1}{\rho c_{t,l}^2} \left[ \left( \chi_{t,l}^{\text{rel}} \right)^2 + 2\chi_{t,l}^{\text{rel}} \chi_{t,l}^{\text{res}}(0) \right] < 0$  is always negative, which means with the increase of length scale, the relaxation susceptibility becomes ‘‘more and more negative’’. The relaxation susceptibility is therefore not marginally irrelevant, but renormalization relevant as well.

It is at this point that all of the beautiful theoretical explanations on experimental measurements breaks down. To illustrate this point of view, let us pretend that the renormalization equations, Eqs.(4.19) are

renormalization irrelevant. Therefore, the renormalization equation for relaxation susceptibility has a non-trivial stable fixed point and a trivial stable fixed point:

$$\chi_{t,l}^{\text{rel}}(R) = -2\chi_{t,l}^{\text{res}}(\omega = 0, R) \quad (4.23)$$

$$\chi_{t,l}^{\text{rel}}(R) = 0 \quad (4.24)$$

Eq.(4.23) is the main result to explain the universal shift of sound velocity in glass, if the renormalization equations are marginally irrelevant. The non-trivial stable fixed point, Eq.(4.23) indicates that even if relaxation and resonance susceptibilities are entirely different at microscopic level, at experimental large length scale relaxation susceptibility always flows to  $-2$  of resonance susceptibility with zero-frequency.

However, the truth is, both of relaxation and resonance susceptibilities are renormalization relevant. In that case, one would presumably have to draw the conclusion that the renormalization procedure increases both the (negative) relaxation and resonance susceptibilities. Then we think one would have to conclude that the starting-scale value of resonance susceptibility is considerably smaller even than the experimental value — a surprising and interesting conclusion! Also, the fixed points Eqs.(4.23, 4.24) are actually unstable, which means at experimental length scale, there is no reason that in Eq.(4.23), relaxation susceptibility must equal to  $-2$  of resonance susceptibility (what is more, since relaxation and resonance susceptibilities are both negative, it is impossible to get the “ $-2$ ” relation between two negative quantities); on the other hand, since both of the absolute values of relaxation and resonance susceptibilities increase with the increase of length scale, in Eq.(4.24) the relaxation susceptibility has no reason to reach the unstable fixed point  $\chi_{t,l}^{\text{rel}}(R) = 0$  at experimental length scale  $R$ .

## 4.2 Some Discussions of Sound Velocity Shift

In this section we discuss the temperature dependence of longitudinal and transverse ultrasound velocity  $c_{l,t}(T)$  in relaxation and resonance regimes separately. It is convenient to set up a reference frequency shift  $\Delta\omega(k, T_0)$  at some reference temperature  $T_0$ , then consider phonon frequency shift  $\Delta\omega(k, T)$  at arbitrary temperature  $T$ . Since one can always write phonon frequency shift as  $\Delta\omega(k, T) = k\Delta c_{l,t}(T)$ , we get the relative sound velocity shift as follows:

$$\frac{\Delta c_{l,t}(T) - \Delta c_{l,t}(T_0)}{c_{l,t}} = \frac{\text{Re } \chi_{l,t}(\omega, T) - \text{Re } \chi_{l,t}(\omega, T_0)}{2\rho c_{l,t}^2} \quad (4.25)$$



The behavior of sound velocity shift is different in relaxation and resonance regimes. In resonance regime only resonance susceptibility contributes. The real part resonance susceptibility can be derived by Kramers-Kronig relation from the frequency integral on imaginary resonance susceptibility. Using the assumption that reduced imaginary resonance susceptibility  $\text{Im } \tilde{\chi}_{l,t}^{\text{res}}(\omega, T) = (1 - e^{-\beta\hbar\omega})^{-1} \text{Im } \chi_{l,t}^{\text{res}}(\omega, T)$  is approximately a constant of frequency up to the order of  $\omega_c \sim 10^{15} \text{Hz}$ , and temperatures around 1K[34, 25], we obtain the logarithmic temperature dependence of relative sound velocity shift:

$$\left. \frac{\Delta c_{l,t}(T) - \Delta c_{l,t}(T_0)}{c_{l,t}(T_0)} \right|_{\text{res}} = \frac{2}{2\pi\rho c_{l,t}^2} \mathcal{P} \int_0^\infty \frac{\Omega \left( \text{Im } \chi_{l,t}^{\text{res}}(\Omega, T) - \text{Im } \chi_{l,t}^{\text{res}}(\Omega, T_0) \right)}{\Omega^2 - \omega^2} d\Omega = \mathcal{C}_{l,t} \ln \left( \frac{T}{T_0} \right) \quad (4.26)$$

where  $\mathcal{C}_{l,t} = -\text{Im } \tilde{\chi}_{l,t}^{\text{res}}/2\pi\rho c_{l,t}^2$  is a positive constant proportional to reduced imaginary resonance susceptibility. For the calculations of the above Eq.(4.26), please see Appendix (E) for details. Eq.(4.26) is a multiple-level generalization of TTLS derivation on sound velocity shift[33]. The constant  $\mathcal{C}_{l,t}$  is not the functional of phonon frequency.

Next we discuss sound velocity shift in relaxation regime. It has contributions from real part resonance and relaxation susceptibilities. The real part contribution of resonance susceptibility in relaxation regime is still  $\mathcal{C}_{l,t} \ln(T/T_0)$ . If, we pretend that the fixed point Eq.(4.23) is stable, then from the ‘‘stable’’ fixed point, the relaxation susceptibility equals to  $-2$  of zero-frequency resonance susceptibility at experimental length scale,  $\Delta \text{Re } \chi_{l,t}^{\text{rel}}(\omega, T)/(2\rho c_{l,t}^2) = -2\mathcal{C}_{l,t} \ln(T/T_0)$ . Finally, the sound velocity shift in relaxation regime is

$$\left. \frac{\Delta c_{l,t}(T) - \Delta c_{l,t}(T_0)}{c_{l,t}} \right|_{\text{rel}} = \frac{\Delta \text{Re} \left( \chi_{l,t}^{\text{rel}}(\omega, T) + \chi_{l,t}^{\text{res}}(\omega, T) \right)}{2\rho c_{l,t}^2} = -\mathcal{C}_{l,t} \ln \left( \frac{T}{T_0} \right) \quad (4.27)$$

Summarize Eq.(4.26, 4.27) the slope ratio of temperature dependence of sound velocity shift in relaxation and resonance regimes is given by  $\mathcal{C}_{l,t}^{\text{rel}} : \mathcal{C}_{l,t}^{\text{res}} = -1 : 1$ . Unfortunately, due to the increasing behavior of relaxation and resonance susceptibilities, the fixed point Eq.(4.23) is unstable. One can never reach the conclusion that relaxation susceptibility equals to  $-2$  of resonance susceptibility at zero-frequency. In fact, since both of the relaxation and resonance susceptibilities are always negative, it is impossible that they have opposite signs at experimental length scale.

### 4.3 Dielectric Shift as the Function of Temperature

The low-temperature dielectric constant is a monotonically decreasing function of temperature in crystalline material[59], while in glass materials, the low-temperature dielectric constant decreases as the increase of temperature first, then increases (see Fig.4.6). In the language of TTLS model, our conjecture is that in glass

material TTLS scatter propagating photons, resulting in the change of photon speed (dielectric constant) as the function of temperature. It is the different influences from relaxation and resonance processes upon photon propagation which gives rise to such kind of anomalous dielectric constant shift. The input electric field frequency of glass dielectric experiment is of order  $f = 500\text{Hz} \sim 50\text{kHz}$ [15], with the wavelength  $\lambda = 6 \times 10^3\text{m} \sim 6 \times 10^5\text{m}$ , much greater than the experimental sample length  $L$ . Another experiment by M. v. Schickfus[14], however, has the input frequency  $f = 10\text{GHz}$ , corresponding to wavelength  $\lambda = 3 \times 10^{-2}\text{m}$  smaller than sample length. We still consider a block of glass with the size much greater than atomic distance  $L \gg a$ . Expanding the glass total electro-magnetic Hamiltonian  $\hat{H}_{EM}^{\text{tot}}$  in orders of electric field in long wavelength limit ( $\lambda \gg a$ ), we obtain

$$\hat{H}_{EM}^{\text{tot}} = \hat{H}_{EM;0}^{\text{tot}} + \int d^3x \sum_i E_i(\vec{x}) \hat{P}_i^{\text{tot}}(\vec{x}) + \mathcal{O}(E^2) \quad (4.28)$$

where the vector operator  $\hat{P}^{\text{tot}}(\vec{x})$  is defined by

$$\hat{P}_i^{\text{tot}}(\vec{x}) = \frac{\delta \hat{H}_{EM}^{\text{tot}}}{\delta E_i(\vec{x})} \quad (4.29)$$

By taking operator derivative  $\hat{P}_i^{\text{tot}}$  with respect to electric field we further define the susceptibility

$$\chi_{ij}^{\text{tot}}(\vec{x} - \vec{x}', t - t') = \frac{\delta \langle \hat{P}_i^{\text{tot}} \rangle(\vec{x}, t)}{\delta E_j(\vec{x}', t')} \quad (4.30)$$

Again in the above definition the average operator stands for thermal and quantum averages, with the temperature  $\beta = (k_B T)^{-1}$ . Let us separate the Hamiltonian  $\hat{H}_{EM}^{\text{tot}}$  into purely electric part  $\hat{H}_{EM}^{\text{el}}$  and dielectric Hamiltonian  $\hat{H}_{EM}^{\text{non}}$ . The electric part  $\hat{H}_{EM}^{\text{el}}$  can be represented by free electro-magnetic fields:

$$\hat{H}_{EM}^{\text{el}} = \int d^3x \sum_i \left( \frac{\epsilon}{2} E_i(\vec{x}) E_i(\vec{x}) + \frac{1}{2\mu} B_i(\vec{x}) B_i(\vec{x}) \right) \quad (4.31)$$

We further define dielectric vector operator  $\hat{P}_i^{\text{non}}(\vec{x})$  and dielectric susceptibility  $\chi_{ij}^{\text{non}}(\vec{x} - \vec{x}', t - t')$  which comes from the dielectric Hamiltonian  $\hat{H}_{EM}^{\text{non}}$ ,

$$\begin{aligned} \hat{H}_{EM}^{\text{non}} &= \hat{H}_{EM;0}^{\text{non}} + \int d^3x \sum_i E_i(\vec{x}) \hat{P}_i^{\text{non}}(\vec{x}) + \mathcal{O}(E^2) \\ \hat{P}_i^{\text{non}}(\vec{x}) &= \frac{\delta \hat{H}_{EM}^{\text{non}}}{\delta E_i(\vec{x})} \quad \chi_{ij}^{\text{non}}(\vec{x} - \vec{x}', t - t') = \frac{\delta \langle \hat{P}_i^{\text{non}} \rangle(\vec{x}, t)}{\delta E_j(\vec{x}', t')} \end{aligned} \quad (4.32)$$

In the following discussions we will use  $\hat{H}_{EM;0}$ ,  $\chi_{ij}$  and  $\hat{P}_i$  to stand for dielectric Hamiltonian, susceptibility and vector operators  $\hat{H}_{EM;0}^{\text{non}}$ ,  $\chi_{ij}^{\text{non}}$  and  $\hat{P}_i^{\text{non}}$ , while we use  $\hat{H}_{EM}^{\text{el}}$ ,  $\chi_{ij}^{\text{el}}$  and  $\hat{P}_i^{\text{el}}$  to stand for the purely electric Hamiltonian, susceptibility and vector operator. The dielectric vector operator  $\hat{P}_i$  is not electric dipole moment operators we usually use in dielectric materials. In fact, let's consider a dielectric system with electronic dipole moments  $\hat{p}_i(\vec{x}) = q(\vec{x})l_i(\vec{x})$  embedded in it. The total electro-magnetic Hamiltonian of glass is:

$$\hat{H}_{EM}^{\text{tot}} = \hat{H}_{EM;0}^{\text{tot}} + \int d^3x \sum_i \left( \frac{1}{2} \epsilon E_i(\vec{x}) E_i(\vec{x}) + \frac{1}{2\mu} B_i(\vec{x}) B_i(\vec{x}) - E_i(\vec{x}) \hat{p}_i(\vec{x}) \right) \quad (4.33)$$

Compare Eq.(4.32) and Eq.(4.33), the operator  $\hat{P}_i(\vec{x})$  is the negative of electronic dipole moments:  $\hat{P}_i(\vec{x}) = -\hat{p}_i(\vec{x})$ . To calculate the space-averaged dielectric susceptibility  $\chi_{ij}(\omega) = \frac{1}{L^3} \int d^3x d^3x' \chi_{ij}(\vec{x} - \vec{x}', \omega)$ , let's denote  $|m\rangle$  and  $E_m$  to be the  $m$ -th eigenstate and eigenvalue of dielectric Hamiltonian  $\hat{H}_{EM;0}$ . Using linear response theory, dielectric susceptibility is given by

$$\begin{aligned} \chi_{ij}(\omega) &= \frac{1}{1 - i\omega\tau} \chi_{ij}^{\text{rel}} + \chi_{ij}^{\text{res}}(\omega + i\eta) \\ \chi_{ij}^{\text{rel}} &= \frac{\beta}{L^3} \sum_n \int d^3x d^3x' \left( \sum_m P_n P_m \langle n | \hat{p}_i(\vec{x}) | n \rangle \langle m | \hat{p}_j(\vec{x}') | m \rangle - P_n \langle n | \hat{p}_i(\vec{x}) | n \rangle \langle n | \hat{p}_j(\vec{x}') | n \rangle \right) \\ \chi_{ij}^{\text{res}}(\omega + i\eta) &= \frac{1}{L^3 \hbar} \sum_n \sum_{l \neq n} \int d^3x d^3x' P_n \frac{\langle n | \hat{p}_i(\vec{x}) | l \rangle \langle l | \hat{p}_j(\vec{x}') | n \rangle}{\omega + (E_n - E_l)/\hbar + i\eta} \\ &\quad - \frac{1}{L^3 \hbar} \sum_l \sum_{n \neq l} \int d^3x d^3x' P_l \frac{\langle n | \hat{p}_i(\vec{x}) | l \rangle \langle l | \hat{p}_j(\vec{x}') | n \rangle}{\omega + (E_n - E_l)/\hbar + i\eta} \end{aligned} \quad (4.34)$$

Where  $P_n = e^{-\beta E_n} / \mathcal{Z}$  stands for the distribution function of the  $n$ -th eigenstate.  $\tau$  is the effective multiple-level-system  $\hat{H}_{EM;0}$  relaxation time. Since the dielectric susceptibility must be invariant under SO(3) group transformations, it takes the generic form  $\chi_{ij}(\omega) = \chi(\omega) \delta_{ij}$ . Similar with phonon frequency shift, photon frequency can be shifted by dielectric susceptibility  $\chi(\omega)$ :

$$\frac{\Delta\omega_k}{\omega_k} = \frac{\chi(\omega)}{2\epsilon} \quad (4.35)$$

where the real part frequency shift corresponds to dielectric constant shift, and the imaginary part frequency shift corresponds to dielectric loss  $\alpha$ . Dielectric susceptibility has relaxation and resonance parts to shift

dielectric constant:

$$\begin{aligned}\frac{\Delta\epsilon_r}{\epsilon_r} &= -\frac{\text{Re}(\chi^{\text{res}}(\omega) + \chi^{\text{rel}}(\omega))}{\epsilon} && \text{relaxation regime} \\ \frac{\Delta\epsilon_r}{\epsilon_r} &= -\frac{\text{Re}\chi^{\text{res}}(\omega)}{\epsilon} && \text{resonance regime}\end{aligned}\quad (4.36)$$

To explore the dielectric shift as the functional of temperature, we want to find the temperature dependence of real part dielectric susceptibility in different regimes.

One may realize that the Hamiltonian Eq.(4.33) is incomplete, because electric dipole moments  $\hat{p}_i(\vec{x})$  can interact with each other via  $1/r^3$  dipole-dipole interaction. In fact, we can also derive electric dipole-dipole interaction by virtual photon exchange process:

$$\hat{U} = \sum_{i,j=1}^3 \int d^3x d^3x' \mu_{ij}(\vec{x} - \vec{x}') \hat{p}_i(\vec{x}) \hat{p}_j(\vec{x}') \quad \mu_{ij}(\vec{x} - \vec{x}') = \frac{\delta_{ij} - 3n_i n_j}{8\pi\epsilon|\vec{x} - \vec{x}'|^3} \quad (4.37)$$

where  $n_i$  is the  $i$ -th component of unit vector of  $\vec{x} - \vec{x}'$ . If we combine  $N_0^3$  copies of  $L \times L \times L$  glass unit blocks to form a  $N_0L \times N_0L \times N_0L$  super block, dipole-dipole interaction between unit blocks will affect glass super block dielectric Hamiltonian. In the following discussions we will always use the approximation to replace  $\vec{x}$  by  $\vec{x}_s$  for the center of  $s$ -th unit block, where  $s = 1, 2, \dots, N_0^3$ , and that  $\int_{V(s)} \hat{p}_i(\vec{x}) d^3x = p_i^{(s)}$  is the uniform electric dipole moment of the  $s$ -th block. Also, we use  $\vec{E}^{(s)}(t)$  to denote the uniform electric field of the  $s$ -th block. With the presence of external electric field, the glass super block dielectric Hamiltonian is given by

$$\hat{H}_{EM} = \sum_{s=1}^{N_0^3} \left( \hat{H}_{EM;0}^{(s)} - \sum_{i=1}^3 E_i^{(s)}(t) p_i^{(s)} \right) + \sum_{s \neq s'}^{N_0^3} \sum_{ij} \mu_{ij}^{(ss')} \hat{p}_i^{(s)} \hat{p}_j^{(s')} \quad (4.38)$$

From now on we assume the uniform dipole moments'  $\hat{p}_i^{(s)}$  correlation function (dielectric susceptibility) are diagonal in spacial coordinates in glass:  $\chi_{ij}^{(ss')} = \frac{1}{L^3} \langle \hat{p}_i^{(s)} \hat{p}_j^{(s')} \rangle = \chi_{ij} \delta_{ss'}$ . Please note that different from phonon field, electric field is not a collection of real particle oscillations. Therefore the relative positions between differen blocks  $\vec{x}_s - \vec{x}'_s$  will not be modified by external electric field. Hence the dipole-dipole interaction coefficient  $\mu_{ij}(\vec{x} - \vec{x}')$  keeps unchanged under the presence of external field, and the super-block electric dipole moment  $\hat{p}_i^{\text{super}}$  is the direct summation of unit block dipole moments:

$$\hat{p}_i^{\text{super}} = \sum_{s=1}^{N_0^3} e^{i\vec{k} \cdot \vec{x}_s} \hat{p}_i^{(s)} \quad (4.39)$$

Let's denote  $|n^*\rangle$  and  $E_n^*$  to be the  $n$ -th eigenstate and eigenvalue for super block unperturbed Hamiltonian  $\sum_{s=1}^{N_0^3} \hat{H}_{EM;0}^{(s)} + \sum_{s \neq s'}^{N_0^3} \sum_{ij} \mu_{ij}^{(ss')} \hat{p}_i^{(s)} \hat{p}_j^{(s')}$ . With the definition of dielectric susceptibility Eq.(4.32), super block dielectric susceptibility is given by

$$\begin{aligned} \chi_{ij}^{\text{super}}(\omega) &= \frac{1}{(N_0 L)^3} \frac{\beta}{1 - i\omega\tau} \left( \sum_{n^* m^*} \frac{e^{-\beta(E_n^* + E_m^*)}}{\mathcal{Z}^{*2}} \langle n^* | \hat{p}_{i,cc}^{\text{super}} | n^* \rangle \langle m^* | \hat{p}_j^{\text{super}} | m^* \rangle \right. \\ &\quad \left. - \sum_{n^*} \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \langle n^* | \hat{p}_{i,cc}^{\text{super}} | n^* \rangle \langle n^* | \hat{p}_j^{\text{super}} | n^* \rangle \right) \\ &\quad + \frac{1}{(N_0 L)^3} \frac{2}{\hbar} \sum_{n^* l^*} \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \frac{(E_l^* - E_n^*)/\hbar}{(\omega + i\eta)^2 - (E_l^* - E_n^*)^2/\hbar^2} \langle l^* | \hat{p}_{i,cc}^{\text{super}} | n^* \rangle \langle n^* | \hat{p}_j^{\text{super}} | l^* \rangle \end{aligned} \quad (4.40)$$

where  $\hat{p}_{i,cc}^{\text{super}}$  stands for the complex conjugate of  $\hat{p}_i^{\text{super}}$ . The first and second lines of Eq.(4.40) are super block relaxation and resonance susceptibilities. Next we want to sep up the relation between microscopic and macroscopic dielectric susceptibilities. Since the unit and super blocks' length scales differ by a factor of  $N_0$ , repeating this renormalization procedure will carry out experimental length scale susceptibility. We still choose starting small length scale  $L_1 \sim 50\text{\AA}$ . In the  $n$ -th step renormalization, the unit and super block length scales are  $L_n$  and  $N_0 L_n$ . We begin with bare Hamiltonian  $\sum_{s=1}^{N_0^3} \hat{H}_{EM;0}^{(s)}$ , eigenstates  $|n\rangle = \prod_{s=1}^{N_0^3} |n^{(s)}\rangle$  and eigenvalues  $E_n = \sum_{s=1}^{N_0^3} E_n^{(s)}$ . We assume electric dipole-dipole interaction  $\hat{U}$  is relatively weak compared to  $\sum_{s=1}^{N_0^3} \hat{H}_{EM;0}^{(s)}$ , so it can be treated as perturbation. The relations between  $|n^*\rangle$ ,  $E_n^*$  and  $|n\rangle$ ,  $E_n$  are

$$|n^*\rangle = |n\rangle + \sum_{m \neq n} \frac{\langle m | \hat{U} | n \rangle}{E_n - E_m} |m\rangle + \mathcal{O}(U^2) \quad E_n^* = E_n + \langle n | U | n \rangle + \sum_{m \neq n} \frac{|\langle m | \hat{U} | n \rangle|^2}{E_n - E_m} |m\rangle + \mathcal{O}(U^2) \quad (4.41)$$

One can expand super block dielectric susceptibility up to the first orders of  $\hat{U}$  to rewrite super block susceptibility in terms of unit block susceptibility:

$$\begin{aligned} \chi_{ij}^{\text{super}}(\omega) &= \frac{1}{1 - i\omega\tau} \chi_{ij}^{\text{super rel}} + \chi_{ij}^{\text{super res}}(\omega + i\eta) \\ &= \frac{1}{1 - i\omega\tau} \left\{ \chi_{ij}^{\text{rel}} - \frac{L_n^3}{N_0^3} \left[ - \sum_{ab} \sum_{ss'} \mu_{ab}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \right] (\chi_{ia}^{\text{rel}} \chi_{bj}^{\text{rel}} + 2\chi_{ia}^{\text{rel}} \chi_{bj}^{\text{res}}(0)) \right\} \\ &\quad + \chi_{ij}^{\text{res}}(\omega + i\eta) - \frac{L_n^3}{N_0^3} \left[ - \sum_{ab} \sum_{ss'} \mu_{ab}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \right] \chi_{ia}^{\text{res}}(\omega + i\eta) \chi_{bj}^{\text{res}}(\omega + i\eta) \end{aligned} \quad (4.42)$$

where  $\frac{1}{1 - i\omega\tau} \chi_{ij}^{\text{super rel}}$  and  $\chi_{ij}^{\text{super res}}(\omega + i\eta)$  are super block relaxation and resonance dielectric susceptibilities.

Applying symmetry property of dielectric susceptibility  $\chi_{ij} = \chi \delta_{ij}$  the renormalization equations can be

further simplified as:

$$\begin{aligned}\chi^{\text{super rel}} &= \chi^{\text{rel}} - \frac{1}{3\epsilon}\chi^{\text{rel}} [\chi^{\text{rel}} + 2\chi^{\text{res}}(0)] \\ \chi^{\text{super res}}(\omega + i\eta) &= \chi^{\text{res}}(\omega + i\eta) - \frac{1}{3\epsilon} [\chi^{\text{res}}(\omega + i\eta)]^2\end{aligned}\quad (4.43)$$

Eq.(4.43) is very similar to the renormalization equations of non-elastic stress-stress susceptibility. We now examine the implications of these renormalization equations. At the first glance, it seems that the dielectric resonance susceptibility presents usual marginally renormalization irrelevant behavior with the increase of length scale: by repeating renormalization procedure for the modulus of resonance susceptibility from starting small length scale  $L_1$  to experimental length scale  $R$  we get logarithmic length scale dependence as follows

$$\frac{1}{|\chi^{\text{res}}(\omega + i\eta, R)|} = \frac{1}{3\epsilon} \ln\left(\frac{R}{L_1}\right) + \frac{1}{|\chi^{\text{res}}(\omega + i\eta, L_1)|}\quad (4.44)$$

On the other hand, there is a fixed point in the renormalization equation of relaxation susceptibility in Eqs.(4.43):

$$\chi^{\text{rel}}(R) = -2\chi^{\text{res}}(\omega = 0, R)\quad (4.45)$$

The ‘‘experimental length scale  $R$ ’’ is the minimum of sample length scale  $L$  and input electric field wavelength  $\lambda$ :  $R = \min(L, \lambda)$ . In the problem of sound velocity shift,  $L > \lambda$ . In this dielectric shift problem, for input frequency  $480\text{Hz} < f < 50\text{kHz}$ [15] we have  $R = L < \lambda$ , while for input frequency  $f = 10\text{GHz}$  by M. v. Schickfus[14], we have  $L > \lambda = R$ .

However, if we stare at the definitions of relaxation and resonance susceptibilities in Eq.(4.34), we find that they are not positive, but are negative quantities. First of all, the resonance susceptibility is negative. For example, let us choose  $\omega + i\eta = 0$ , and let  $i = j$  in resonance susceptibility, to consider  $\chi_{ij}^{\text{res}}(\omega + i\eta = 0)$ ,

$$\chi_{ij}^{\text{res}}(\omega + i\eta = 0) = \frac{1}{L^3} \sum_{nl} \left( \frac{P_n - P_l}{E_n - E_l} \right) |\langle n|\hat{p}_i|l\rangle|^2 < 0\quad (4.46)$$

The resonance susceptibility  $\chi_{ij}^{\text{res}}(\omega + i\eta = 0)$  is negative mainly because  $P_n < P_l$  for arbitrary pair of levels  $n, m$  with  $E_n > E_l$ . Because of the negativity of resonance susceptibility, the renormalization equation of resonance susceptibility is actually not marginally irrelevant, but renormalization relevant (see the second equation of Eqs.(4.43)).

On the other hand, the relaxation susceptibility  $\chi_{ij}^{\text{rel}}$  is negative as well. To prove this result let us define  $\sum_n P_n \langle n | \hat{p}_i | n \rangle = \bar{p}_i$  to be the ‘‘average value of dipole moment operator  $\hat{p}_i$ ’’, and define  $\sum_n P_n \langle n | \hat{p}_i | n \rangle^2 = (\bar{p}_i^2)$  to be the ‘‘average value of the square of electric dipole moment operator  $\hat{p}_i$ ’’. For simplicity we still let  $i = j$  to consider the relaxation susceptibility. The relaxation susceptibility  $\chi_{ii}^{\text{rel}}$  is rewritten as follows,

$$\begin{aligned}
\chi_{ii}^{\text{rel}} &= \frac{\beta}{L^3} \sum_n \int d^3x d^3x' \left( \sum_m P_n P_m \langle n | \hat{p}_i(\vec{x}) | n \rangle \langle m | \hat{p}_i(\vec{x}') | m \rangle - P_n \langle n | \hat{p}_i(\vec{x}) | n \rangle \langle n | \hat{p}_i(\vec{x}') | n \rangle \right) \\
&= \frac{\beta}{L^3} \left( \sum_n \sum_m P_n P_m \langle n | \hat{p}_i | n \rangle \langle m | \hat{p}_i | m \rangle - \sum_n P_n \langle n | \hat{p}_i | n \rangle \langle n | \hat{p}_i | n \rangle \right) \\
&= \frac{\beta}{L^3} \left( (\bar{p}_i)^2 - (\bar{p}_i^2) \right) < 0
\end{aligned} \tag{4.47}$$

Therefore, the relaxation susceptibility is always negative as well. If we look back to the first renormalization equation of Eqs.(4.43), the linear term  $-\frac{1}{3\epsilon} \left[ (\chi^{\text{rel}})^2 + 2\chi^{\text{rel}}\chi^{\text{res}}(0) \right] < 0$  is always negative, which means with the increase of length scale, the relaxation susceptibility becomes ‘‘more and more negative’’. The relaxation susceptibility is therefore not marginally irrelevant, but renormalization relevant as well.

Similar with the theoretical explanation on universal shift of sound velocity in glass, all of the theoretical explanations on experimental measurements of dielectric constant break down. To illustrate this point of view, let us pretend that the renormalization equations, Eqs.(4.43) are renormalization irrelevant. Therefore, the renormalization equation for relaxation susceptibility has a non-trivial stable fixed point which was shown in Eq.(4.45).

Eq.(4.45) is the main result to explain the universal shift of dielectric constant in glass, if the renormalization equations are marginally irrelevant. The non-trivial stable fixed point, Eq.(4.45) indicates that even if relaxation and resonance susceptibilities are entirely different at microscopic level, at experimental large length scale relaxation susceptibility always flows to  $-2$  of resonance susceptibility with zero-frequency.

However, the truth is, both of relaxation and resonance susceptibilities are renormalization relevant. Therefore the renormalization procedure increases both the (negative) relaxation and resonance susceptibilities, which means the starting-scale value of resonance susceptibility is considerably smaller even than the experimental value. Also since relaxation and resonance susceptibilities are both negative, it is impossible to get the ‘‘ $-2$ ’’ relation between two negative quantities in the fixed point Eq.(4.45).

Since dielectric susceptibility is functional of temperature, it is convenient to set up a reference dielectric shift  $\Delta\epsilon_r(T_0)$  at some reference temperature  $T_0$ . The relative shift of dielectric constant at temperature  $T$  is

$$\frac{\Delta\epsilon_r(T) - \Delta\epsilon_r(T_0)}{\epsilon_r} = - \frac{\text{Re}\chi(\omega, T) - \text{Re}\chi(\omega, T_0)}{\epsilon} \tag{4.48}$$

In resonance regime, the real part of dielectric resonance susceptibility can be obtained by integrating over imaginary part of dielectric susceptibility via Kramers-Kronig relation. Again we use the assumption that reduced imaginary resonance susceptibility  $\text{Im } \tilde{\chi}^{\text{res}}(\omega, T) = (1 - e^{-\beta\hbar\omega})^{-1} \text{Im } \chi^{\text{res}}(\omega, T)$  is approximately the constant of frequency and temperature around 1K[8], we obtain the logarithmic temperature dependence of relative dielectric constant shift:

$$\left. \frac{\Delta\epsilon_r(T) - \Delta\epsilon_r(T_0)}{\epsilon_r(T_0)} \right|_{\text{res}} = -\frac{2}{\pi\epsilon} \mathcal{P} \int_0^\infty \frac{\Omega (\text{Im } \chi^{\text{res}}(\Omega, T) - \text{Im } \chi^{\text{res}}(\Omega, T_0))}{\Omega^2 - \omega^2} d\Omega = -\mathcal{C} \ln\left(\frac{T}{T_0}\right) \quad (4.49)$$

where  $\mathcal{C} = -\text{Im } \tilde{\chi}^{\text{res}}/\pi\epsilon$  is a positive constant proportional to the reduced imaginary resonance susceptibility.  $\mathcal{C}$  is independent of frequency  $\omega$ .

Next we discuss dielectric constant shift in relaxation regime. It has contributions from real part resonance and relaxation susceptibilities. The real part contribution of resonance susceptibility in relaxation regime is still  $\mathcal{C} \ln(T/T_0)$ . If, we pretend that the fixed point Eq.(4.45) is stable, then from the ‘‘stable’’ fixed point, the relaxation susceptibility equals to  $-2$  of zero-frequency resonance susceptibility at experimental length scale,  $\Delta\text{Re } \chi^{\text{rel}}(\omega, T)/\epsilon = 2\mathcal{C} \ln(T/T_0)$ . Finally, the dielectric constant shift in relaxation regime is

$$\left. \frac{\Delta\epsilon_r(T) - \Delta\epsilon_r(T_0)}{\epsilon_r} \right|_{\text{rel}} = -\frac{\Delta\text{Re}(\chi^{\text{rel}}(\omega, T) + \chi^{\text{res}}(\omega, T))}{\epsilon} = \mathcal{C} \ln\left(\frac{T}{T_0}\right) \quad (4.50)$$

Summarize Eq.(4.49, 4.50) the slope ratio of temperature dependence of dielectric constant shift in relaxation and resonance regimes is given by  $\mathcal{C}^{\text{rel}} : \mathcal{C}^{\text{res}} = 1 : -1$ . Unfortunately, due to the increasing behavior of relaxation and resonance susceptibilities, the fixed point Eq.(4.45) is unstable. One can never conclude that relaxation susceptibility equals to  $-2$  of resonance susceptibility at zero-frequency. In fact, since both of the relaxation and resonance susceptibilities are always negative, it is impossible that they have opposite signs at experimental length scale. Compare the slope ratio of dielectric constant shift with that of sound velocity shift, the negative sign appears in the definition of electric dipole moment.



# Chapter 5

## Low Temperature Insulating Glass Mechanical Avalanche Problem

### 5.1 The Set up of Avalanche Problem

The glass mechanical avalanche phenomena is referred to the glass stress-strain curve which presents a steep drop to a lower value at certain critical external strain when avalanche happens[23]. The purpose of this chapter is to develop a tentative microscopic field theory to investigate such mechanical property of three-dimensional insulating glass under the deformation of external static, uniform strain. The reader should be aware that this is the first time to apply “generic coupled block model” in glass mechanical avalanche problem. Therefore our purpose is not to solve the entire glass avalanche problem from microscopic point of view; instead we want to provide some first-step results for future people to continue studying this problem. In the following renormalization analysis of Eq.(5.22), we will find that since the non-elastic susceptibility stays negative throughout the entire renormalization procedure, it is impossible to find positive-negative transitions in non-elastic susceptibility. We hope to provide some help for future people to further explore glass avalanche problems.

As we will see later, the effective starting microscopic length scale of our real space renormalization procedure is of order  $\sim 50\text{\AA}$ , corresponding to the characteristic thermal phonon wavelength with the temperature of order 50K. Our explanation is only valid below this temperature. However, at least to the author’s knowledge, all of glass avalanche experiments are taken under room temperatures or glass transition temperatures[21, 43, 44, 39, 40] ( $T \sim 300\text{K}$ ). We hope more experiments on such mechanical properties of glass could be taken at low-temperatures below 50K.

Let’s consider a block of glass material. With the slowly increasing external strain the bulk glass behaves elastically until it reaches critical strain value. The stress ( $\mathbf{T}$ ) v.s. strain ( $\mathbf{e}$ ) curve shows a steep drop. A much more convenient quantity we consider is the mechanical stress-stress susceptibility  $\chi_{ijkl}(\mathbf{e}) = \delta T_{ij} / \delta e_{kl}$ . At critical external strain field when irreversible process happens, stress-stress susceptibility presents an abrupt positive-negative transition, which is shown in Fig.5.1 as follows:

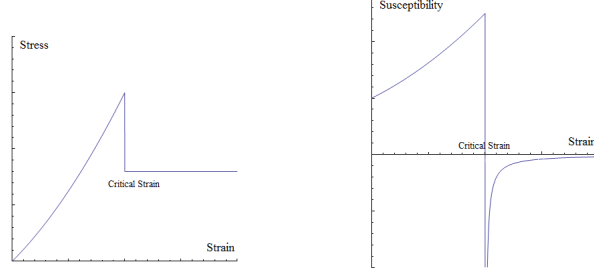


Figure 5.1: As an illustration of stress-strain curve, the left picture shows a steep drop of stress. As an illustration of susceptibility-strain curve, the right picture shows a positive-negative susceptibility transition, where susceptibility is the first order derivative of stress with respect to external strain field.

The purpose of this chapter is to understand such mechanical property of three-dimensional insulating glass under the deformation of external static, uniform strain. Since we do not take conducting electrons into consideration, our model only applies for insulating glass. Further considerations regarding conducting electron Hamiltonian, electron-phonon coupling and electron-stress tensor coupling are required to explore the ductility of metallic glass. In this chapter our main goal is to prove the existence of such mechanical susceptibility positive-negative transition. We start our problem by considering a block of glass with the length scale  $L$  much greater than the atomic distance  $a \sim 10\text{\AA}$ . Please note, that in this section we have not put in external static uniform external strain field yet. We further define the elastic strain field  $e_{ij}(\vec{x})$  which is the spacial derivative of matter displacement  $\vec{u}(\vec{x})$  at position  $\vec{x}$ :  $e_{ij}(\vec{x}) = \frac{1}{2} \left( \frac{\partial u_i(\vec{x})}{\partial x_j} + \frac{\partial u_j(\vec{x})}{\partial x_i} \right)$ . We write general glass Hamiltonian as  $\hat{H}^{\text{tot}}$ , and expand it in orders of intrinsic elastic strain field  $e_{ij}$  in long wavelength limit ( $\lambda \gg a$ ):

$$\hat{H}^{\text{tot}} = \hat{H}_0^{\text{tot}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{tot}}(\vec{x}) + \mathcal{O}(e_{ij}^2) \quad (5.1)$$

the coefficient of first order expansion is stress tensor  $\hat{T}_{ij}^{\text{tot}}(\vec{x})$ , defined by the derivative of Hamiltonian with respect to intrinsic phonon strain field

$$\hat{T}_{ij}^{\text{tot}}(\vec{x}) = \frac{\delta \hat{H}^{\text{tot}}}{\delta e_{ij}(\vec{x})} \quad (5.2)$$

The most important quantity of this thesis, stress-stress susceptibility  $\chi_{ijkl}^{\text{tot}}$  is defined by taking derivative on stress tensor  $\hat{T}_{ij}^{\text{tot}}$  with respect to intrinsic phonon strain field  $e_{kl}(\vec{x})$ . The susceptibility is taken for the glass block much larger than atomic distance:

$$\chi_{ijkl}^{\text{tot}}(\vec{x} - \vec{x}'; t - t') = \frac{\delta \langle \hat{T}_{ij}^{\text{tot}} \rangle(\vec{x}, t)}{\delta e_{kl}(\vec{x}', t')} \quad (5.3)$$

where the expectation value of stress tensor operator  $\hat{T}_{ij}^{\text{tot}}(\vec{x})$  is functional of time. In Eq.(5.3) the average of  $\langle \hat{T}_{ij}^{\text{tot}} \rangle$  represents thermal and quantum average: for an arbitrary operator  $\hat{A}$ ,  $\langle \hat{A} \rangle = \sum_m \mathcal{Z}^{-1} e^{-\beta E_m} \langle m, t | \hat{A} | m, t \rangle$  with  $|m\rangle$  the eigenbasis of Hamiltonian  $\hat{H}_0$  and  $\mathcal{Z}$  the partition function  $\mathcal{Z} = \sum_m e^{-\beta E_m}$  with temperature  $\beta = (k_B T)^{-1}$ . Susceptibility is also the function of temperature, but for notational simplicity we write  $\chi(\vec{x} - \vec{x}'; t - t'; T)$  as  $\chi(\vec{x} - \vec{x}'; t - t')$ .

In the rest of this chapter it is convenient to separate glass Hamiltonian  $\hat{H}^{\text{tot}}$  into purely elastic part  $\hat{H}^{\text{el}}$  and non-elastic part  $\hat{H}^{\text{non}}$ :  $\hat{H}^{\text{tot}} = \hat{H}^{\text{el}} + \hat{H}^{\text{non}}$ . By taking their first order derivatives with respect to intrinsic phonon strain field, the stress tensor  $\hat{T}_{ij}^{\text{tot}}$  can be separated into elastic and non-elastic stress tensors:  $\hat{T}_{ij}^{\text{tot}}(\vec{x}) = \hat{T}_{ij}^{\text{el}}(\vec{x}) + \hat{T}_{ij}^{\text{non}}(\vec{x})$ . Similarly, the elastic and full non-elastic stress-stress susceptibilities are the corresponding stress tensors' derivatives:

$$\chi_{ijkl}^{\text{tot}}(\vec{k}, \omega) = \chi_{ijkl}^{\text{el}}(\vec{k}, \omega) + \chi_{ijkl}^{\text{non}}(\vec{k}, \omega) \quad (5.4)$$

The purpose of this chapter is to prove that for certain critical external strain field  $e_{ij}$ , the positive stress-stress susceptibility Eq.(5.4) suddenly drops to a negative value, leading to the mechanical avalanche behavior of glass. Subtracting elastic part from glass Hamiltonian, the left-over non-elastic Hamiltonian can be expanded in orders of long wavelength intrinsic phonon strain field:

$$\begin{aligned} \hat{H}^{\text{non}} &= \hat{H}_0^{\text{non}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{non}}(\vec{x}) + \mathcal{O}(e_{ij}^2) \\ \hat{T}_{ij}^{\text{non}}(\vec{x}) &= \frac{\delta \hat{H}^{\text{non}}}{\delta e_{ij}(\vec{x})} \\ \chi_{ijkl}^{\text{non}}(\vec{x} - \vec{x}'; t - t') &= \frac{\delta \langle \hat{T}_{ij}^{\text{non}} \rangle(\vec{x}, t)}{\delta e_{kl}(\vec{x}', t')} \end{aligned} \quad (5.5)$$

where for convenience we will use  $\chi_{ijkl}^{\text{non}}(\vec{x} - \vec{x}'; t - t')$  to stand for  $\chi_{ijkl}^{\text{non}}(\vec{x} - \vec{x}'; t - t'; \mathbf{e})$ . In the rest of this chapter we further use  $\hat{H}_0$ ,  $\chi_{ijkl}$  and  $\hat{T}_{ij}$  to represent  $\hat{H}_0^{\text{non}}$ ,  $\chi_{ijkl}^{\text{non}}$  and  $\hat{T}_{ij}^{\text{non}}$ , while we use  $\hat{H}^{\text{el}}$ ,  $\chi_{ijkl}^{\text{el}}$  and  $\hat{T}_{ij}^{\text{el}}$  to represent the elastic Hamiltonian, susceptibility and stress tensor.

We want to explain avalanche under external static strain field deformations. Therefore we focus on DC ( $\omega = 0$ ) non-elastic stress-stress susceptibility  $\lim_{\omega \rightarrow 0} \chi_{ijkl}(\omega)$ . We denote  $|m\rangle$  and  $E_m$  to be the  $m$ -th eigenstate and eigenvalue of unperturbed non-elastic Hamiltonian  $\hat{H}_0$ . The eigenbasis  $|m\rangle$  is a set of generic multiple-level-system. By using linear response theory, we expand the expectation value of stress tensor  $\langle \hat{T}_{ij} \rangle$  up to the first order of  $e_{ij} \hat{T}_{ij}$  to derive non-elastic stress-stress susceptibility. We use the same language as tunneling-two-level-system, that the susceptibility can be expressed in relaxation and resonance susceptibilities. The relaxation susceptibility comes from the energy eigenvalue shift due to the diagonal

matrix elements of perturbation, while the resonance susceptibility comes from the off-diagonal matrix elements of perturbing Hamiltonian. Let's denote  $\tau$  to be effective thermal relaxation time for glass. We use  $\chi_{ijkl}^{\text{rel}}(\omega)$  to stand for relaxation susceptibility, and use  $\chi_{ijkl}^{\text{res}}(\omega)$  to stand for the resonance susceptibility. The susceptibility is always in relaxation regime because  $\omega\tau = 0$  for external static field. Thus both of zero-frequency relaxation and resonance susceptibilities contribute in full non-elastic stress-stress susceptibility. In the rest of this chapter for simplicity let's use  $\chi_{ijkl}$  to stand for  $\lim_{\omega \rightarrow 0} \chi_{ijkl}(\omega)$ , and use  $\chi_{ijkl}^{\text{rel}}$  and  $\chi_{ijkl}^{\text{res}}$  for  $\lim_{\omega \rightarrow 0} \chi_{ijkl}^{\text{rel}}(\omega)$  and  $\lim_{\omega + i\eta \rightarrow 0} \chi_{ijkl}^{\text{res}}(\omega + i\eta)$ . The zero-frequency susceptibility of generic multiple-level-system is given as follows:

$$\begin{aligned}
\chi_{ijkl} &= \chi_{ijkl}^{\text{rel}} + \chi_{ijkl}^{\text{res}} \\
\chi_{ijkl}^{\text{rel}} &= \frac{\beta}{V} \left( \sum_{nm} P_n P_m \langle n | \hat{T}_{ij} | n \rangle \langle m | \hat{T}_{kl} | m \rangle - \sum_n P_n \langle n | \hat{T}_{ij} | n \rangle \langle n | \hat{T}_{kl} | n \rangle \right) \\
\chi_{ijkl}^{\text{res}} &= -\frac{1}{V\hbar} \sum_n \sum_{m \neq n} P_m \frac{\langle n | \hat{T}_{ij} | m \rangle \langle m | \hat{T}_{kl} | n \rangle}{(E_n - E_m)/\hbar + i\eta} + \frac{1}{V\hbar} \sum_m \sum_{n \neq m} P_n \frac{\langle n | \hat{T}_{ij} | m \rangle \langle m | \hat{T}_{kl} | n \rangle}{(E_n - E_m)/\hbar + i\eta} \quad (5.6)
\end{aligned}$$

where  $\int_V \hat{T}_{ij}(\vec{x}) d^3x = \hat{T}_{ij}$  is the uniform stress tensor of this glass block.  $P_n = e^{-\beta E_n} / \mathcal{Z}$  is the  $n$ -th level probability function and  $\mathcal{Z} = \sum_n e^{-\beta E_n}$  is the partition function with temperature  $\beta = (k_B T)^{-1}$ .  $\eta$  is a phenomenological parameter to represent the higher order corrections of full non-elastic stress-stress susceptibility due to the coupling between strain field and non-elastic stress tensor:  $\sum_{ij} e_{ij} \hat{T}_{ij}$ .

Let us stop here for a moment and check the signs of relaxation and resonance susceptibilities. For example, let us check the diagonal matrix element of relaxation susceptibility,  $\chi_{ijij}^{\text{rel}}$  with indices  $(ij) = (kl)$ . The diagonal matrix element of relaxation susceptibility,  $\chi_{ijij}^{\text{rel}}$  is always negative, because we have the relation  $\sum_{nm} P_n P_m \langle n | \hat{T}_{ij} | n \rangle \langle m | \hat{T}_{ij} | m \rangle < \sum_n P_n |\langle n | \hat{T}_{ij} | n \rangle|^2$ . On the other hand the diagonal matrix element of resonance susceptibility  $\chi_{ijij}^{\text{res}}(\eta = 0)$  with indices  $(ij) = (kl)$  is negative as well, because  $P_n < P_m$  for arbitrary pair of energy levels  $n, m$  with  $E_n > E_m$ . This negative property of relaxation and resonance susceptibilities will be very useful in later discussions of renormalization equation of non-elastic susceptibility.

Next we consider elastic stress-stress susceptibility. The elastic Hamiltonian  $\hat{H}^{\text{el}}$  can be represented by phonon creation-annihilation operators

$$\hat{H}^{\text{el}} = \sum_{k\alpha} \hbar\omega_{k\alpha} \left( \hat{a}_{k\alpha}^\dagger \hat{a}_{k\alpha} + \frac{1}{2} \right) \quad (5.7)$$

where  $\alpha = l, t$  is phonon polarization, i.e., longitudinal and transverse phonons. The elastic complex response

function is therefore given by

$$\chi_{ij,kl}^{\text{el}}(\vec{k}, \omega) = \frac{\delta^2 \langle \hat{H}^{\text{el}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{el}}(\vec{x}) \rangle}{\delta e_{ij} \delta e_{kl}}(\vec{k}, \omega) \quad (5.8)$$

Please note, that in the above definitions of glass elastic susceptibility, the glass elastic Hamiltonian's expectation value is defined by the glass elastic Hamiltonian  $\hat{H}^{\text{el}}$  plus the time-dependent perturbation  $\int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{el}}(\vec{x})$ :  $\hat{H}^{\text{el}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{el}}(\vec{x})$ ; the glass elastic stress response  $\langle \hat{T}_{ij}^{\text{el}}(\vec{x}) \rangle(\vec{x}, t)$  is also defined by using the glass elastic Hamiltonian plus the external time-dependent perturbations:  $\hat{H}^{\text{el}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{el}}(\vec{x})$ .

From the above definitions of elastic and non-elastic glass susceptibilities, at the static limit they are by definition negative. The elastic susceptibility at static limit is given by

$$\chi_{ijkl}^{\text{el}} = -(\rho c_l^2 - 2\rho c_t^2) \delta_{ij} \delta_{kl} - \rho c_t^2 (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) \quad (5.9)$$

The above result seems to be negative compared to the ‘‘elastic constant’’ in a standard elasticity textbook:  $(\rho c_l^2 - 2\rho c_t^2) \delta_{ij} \delta_{kl} + \rho c_t^2 (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk})$ . This is because: in the standard elasticity textbook, one usually defines the ‘‘elastic constant’’ through the definition  $\chi_{ijkl}^{\text{elastic constant}} = \delta^2 \langle \hat{H}^{\text{el}} \rangle / \delta e_{ij} \delta e_{kl}$ , but the Hamiltonian  $\hat{H}^{\text{el}}$  here is the elastic part of glass Hamiltonian which does not include the time-dependent perturbation  $\int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}^{\text{el}}(\vec{x})$ . Therefore our definition of elastic susceptibility differs by a negative sign compared to the standard elastic constant in the standard textbook.

## 5.2 Virtual Phonon Exchange Interactions

The previous problem is within single-block considerations. If we combine a set of such single-blocks together, the interaction between them will be taken into glass Hamiltonian. Since the stress-strain coupling  $e_{ij} \hat{T}_{ij}$  contains phonon strain field  $e_{ij}$ , allowing virtual phonons to exchange will give rise to an effective RKKY-type interaction between different blocks via stress tensor products:

$$\hat{V} = \int d^3x d^3x' \sum_{ijkl} \Lambda_{ijkl}(\vec{x} - \vec{x}') \hat{T}_{ij}(\vec{x}) \hat{T}_{kl}(\vec{x}') \quad (5.10)$$

where the coefficient  $\Lambda_{ijkl}(\vec{x} - \vec{x}')$  was discussed in chapter 2. In the rest of this chapter we still use the approximation to replace  $\vec{x} - \vec{x}'$  by  $\vec{x}_s - \vec{x}_{s'}$  for the  $s$ -th and  $s'$ -th blocks, in which  $\vec{x}_s$  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 stress tensor of the  $s$ -th block. From this definition

the uniform stress tensor operator  $\hat{T}_{ij}^{(s)}$  is volume proportional extensive quantity. The non-elastic part of super block Hamiltonian without external strain field is given by

$$\hat{H}^{\text{super}} = \sum_{s=1}^{N_0^3} \hat{H}_0^{(s)} + \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \quad (5.11)$$

Again, we apply the assumption: to assume that the correlation function of block uniform stress tensors  $\hat{T}_{ij}^{(s)}$  are diagonal in spacial coordinates:  $\chi_{ijkl}^{(ss')} = \frac{1}{L^3} \langle \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \rangle = \chi_{ijkl} \delta_{ss'}$ .

### 5.3 Full Glass Hamiltonian with the Presence of External Static, Uniform Strain field

In this section we begin to put in external static, uniform strain field and consider glass super block Hamiltonian affected by external strain field  $\mathbf{e}(\vec{x}, t)$ . Please note that we have defined non-elastic stress tensor and non-elastic stress-stress susceptibility with the help of intrinsic phonon strain field, in this section  $\mathbf{e}(\vec{x}, t)$  stands for the external real phonon field. Because the purpose of this thesis is to consider avalanche problem under static uniform external strain field, we denote the external strain as  $\mathbf{e}(\vec{x}, t) = \mathbf{e}$  on an isotropic (spherical) glass with radius  $r$ . As the simplest case, we consider the static strain as  $e_{xx} = e$ ,  $e_{yy} = e_{zz} = e_{xy} = e_{yz} = e_{zx} = 0$ . For other kinds of external strain  $\mathbf{e} = e_{ij}$ , similar avalanche behaviors could be found as well. The spherical glass is deformed to be an ellipsoid. The  $xy$  and  $xz$  plane cross sections are ellipses with eccentricity  $\epsilon = \frac{\sqrt{e^2+2e}}{(1+e)}$  while the  $yz$  cross section is circular.

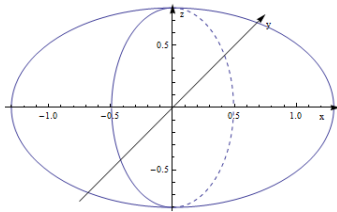


Figure 5.2: An isotropic (spherical) glass deformed by strain  $e_{xx} = e$  to become an ellipsoid.

There are a couple of terms appear in glass Hamiltonian with the turning on of external strain field  $\mathbf{e}$ . First, non-elastic stress tensor operators  $\hat{T}_{ij}^{(s)}$  might be changed for  $\Delta \hat{T}_{ij}^{(s)}$  by external strain field. We further define new single-block stress tensor  $\hat{T}_{ij}^{(s)}(\mathbf{e})$  as follows

$$\hat{T}_{ij}^{(s)}(\mathbf{e}) = \hat{T}_{ij}^{(s)} + \Delta \hat{T}_{ij}^{(s)} = \frac{\delta \hat{H}^{(s)}(\mathbf{e})}{\delta e_{ij}^{(s)}} \quad (5.12)$$

which means the new quantity  $\hat{T}_{ij}^{(s)}(\mathbf{e})$  is non-elastic stress tensor under the presence of external strain  $\mathbf{e}$ . Such strain field dependent property of  $\hat{T}_{ij}^{(s)}(\mathbf{e})$  comes from the nonlinear strain field dependence of non-elastic Hamiltonian. Thus the strain-stress coupling term is given by  $\sum_s \sum_{ij} e_{ij}^{(s)} \hat{T}_{ij}^{(s)}(\mathbf{e})$ , where  $\mathbf{e}$  is external strain. The  $s$ -th unit block full non-elastic susceptibility  $\chi_{ijkl} = V^{-1} \langle \delta^2 \hat{H}^{(s)}(\mathbf{e}) / \delta e_{ij}^{(s)} \delta e_{kl}^{(s)} \rangle$  is given by Eq.(5.6) by replacing  $\hat{T}_{ij}^{(s)}$  with  $\hat{T}_{ij}^{(s)}(\mathbf{e})$ . Virtual phonon exchange process gives non-elastic stress-stress interaction  $\hat{V} = \sum_{ss'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)}(\mathbf{e}) \hat{T}_{kl}^{(s')}(\mathbf{e})$ . In the rest of this thesis we will always write  $\hat{T}_{ij}^{(s)}$  to stand for  $\hat{T}_{ij}^{(s)}(\mathbf{e})$  for simplicity.

There is a second question arising from external strain field: the relative positions of unit blocks  $\vec{x}^{(s)} - \vec{x}^{(s')}$  can be changed by external strain field, resulting in the modification of stress-stress interaction coefficient  $\Lambda_{ijkl}^{(ss')} \rightarrow \Lambda_{ijkl}^{(ss')}(\mathbf{e})$ . Thus the glass super block Hamiltonian is  $\hat{H}^{\text{super}}(\mathbf{e}) = \sum_s \left( \hat{H}_0^{(s)} + \sum_{ij} e_{ij}^{(s)} \hat{T}_{ij}^{(s)} \right) + \sum_{s \neq s'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')}(\mathbf{e}) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}$ . Super block non-elastic stress tensor is defined as  $\hat{T}_{ij}^{\text{super}}(\mathbf{e}) = \delta \hat{H}^{\text{super}}(\mathbf{e}) / \delta e_{ij}$ . Because of the external strain field dependence of  $\Lambda_{ijkl}^{(ss')}(\mathbf{e})$ , an extra term appears in super block stress tensor:

$$\hat{T}_{ij}^{\text{super}} = \sum_s \hat{T}_{ij}^{(s)} + \sum_{ss'} \sum_{abcd} \frac{\delta \Lambda_{abcd}^{(ss')}(\mathbf{e})}{\delta e_{ij}} \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} \quad (5.13)$$

where we use  $\hat{T}_{ij}^{\text{super}}$  to stand for  $\hat{T}_{ij}^{\text{super}}(\mathbf{e})$ .

The super block susceptibility also receives an extra term. To calculate super block susceptibility let us first denote  $|n^*\rangle$  and  $E_n^*$  to be the  $n$ -th eigenstate and eigenvalue of super block unperturbed Hamiltonian  $\hat{H}_0^{\text{super}}(\mathbf{e}) = \sum_s \hat{H}_0^{(s)} + \sum_{ss'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')}(\mathbf{e}) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}$  with perturbation  $\sum_s \sum_{ij} e_{ij}^{(s)} \hat{T}_{ij}^{(s)}$ . By using linear response theory we get super block susceptibility:

$$\begin{aligned} \chi_{ijkl}^{\text{super}} &= \frac{\beta}{(N_0 L)^3} \left( \sum_{nm} P_n^* P_m^* \langle n^* | \sum_s \hat{T}_{ij}^{(s)} | n^* \rangle \langle m^* | \sum_{s'} \hat{T}_{kl}^{(s')} | m^* \rangle - \sum_n P_n^* \langle n^* | \sum_s \hat{T}_{ij}^{(s)} | n^* \rangle \langle n^* | \sum_{s'} \hat{T}_{kl}^{(s')} | n^* \rangle \right) \\ &- \frac{1}{(N_0 L)^3 \hbar} \sum_{nm} (P_m^* - P_n^*) \frac{\langle n^* | \sum_s \hat{T}_{ij}^{(s)} | m^* \rangle \langle m^* | \sum_{s'} \hat{T}_{kl}^{(s')} | n^* \rangle}{(E_n^* - E_m^*) / \hbar + i\eta} \\ &+ \frac{1}{(N_0 L)^3} \sum_{abcd} \sum_{ss'} \left\langle \frac{\delta^2 \Lambda_{abcd}^{(ss')}(\mathbf{e})}{\delta e_{ij} \delta e_{kl}} \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} \right\rangle \end{aligned} \quad (5.14)$$

where we use  $\chi_{ijkl}^{\text{super}}$  to stand for  $\chi_{ijkl}^{\text{super}}(\mathbf{e})$ .

## 5.4 Real Space Renormalization for Glass Non-Elastic Susceptibility

In this section our purpose is to find the non-elastic stress-stress susceptibility at experimental large length scale. We want to set up the relation between unit block and super block non-elastic susceptibilities. Since the super block length scale is  $N_0$  times greater than single block length scale, repeating the recursion relation allows to get experimental length scale non-elastic susceptibility. The suggested renormalization procedure starting length scale is, for example,  $L_1 \sim 50\text{\AA}$  according to the argument of D. C. Vural and A. J. Leggett[25]. Since the final result only logarithmically depends on this choice, it will not be sensitive. The effective starting microscopic length scale must be no less than the order of  $\sim 50\text{\AA}$ , corresponding to the characteristic thermal phonon wavelength with the temperature of order 50K. Again, we combine  $N_0^3$  unit blocks with the dimension  $L_n \times L_n \times L_n$  to form the  $n$ -th step super block glass with the dimension  $N_0 L_n \times N_0 L_n \times N_0 L_n$ . These non-interacting unit blocks have the Hamiltonian  $\hat{H}_0 = \sum_{s=1}^{N_0^3} \hat{H}_0^{(s)}$ , eigenstates  $|n\rangle = \prod_{s=1}^{N_0^3} |n^{(s)}\rangle$  and eigenvalues  $E_n = \sum_{s=1}^{N_0^3} E_n^{(s)}$ . We combine them into a super block and turn on non-elastic stress-stress interactions  $\hat{V}(\mathbf{e}) = \sum_{s \neq s'}^{N_0^3} \Lambda_{ijkl}^{(ss')}(\mathbf{e}) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}$ . We assume non-elastic stress-stress interactions  $\hat{V}$  are relatively weak compared to the summation of unit block Hamiltonians  $\hat{H}_0 = \sum_{s=1}^{N_0^3} \hat{H}_0^{(s)}$ , so that the interactions can be treated as a perturbation. If the non-elastic susceptibility decreases logarithmically as the increase of length scale, then that means the non-elastic stress-stress interaction  $\hat{V}$  can be treated as a perturbation at the late stages. The assumption that  $\hat{V}$  can be treated as a perturbation is qualitatively correct. In the last section we define super block eigenstates and eigenvalues to be  $|n^*\rangle$  and  $E_n^*$ . Their relations with  $|n\rangle$  and  $E_n$  are

$$\begin{aligned}
 |n^*\rangle &= |n\rangle + \sum_{p \neq n} \frac{\langle p | \hat{V}(\mathbf{e}) | n \rangle}{E_n - E_p} |p\rangle + \mathcal{O}(V^2) \\
 E_n^* &= E_n + \langle n | \hat{V}(\mathbf{e}) | n \rangle + \sum_{p \neq n} \frac{|\langle p | \hat{V}(\mathbf{e}) | n \rangle|^2}{E_n - E_p} |p\rangle + \mathcal{O}(V^2)
 \end{aligned} \tag{5.15}$$

With the relations in Eq.(5.15) one can expand super block full non-elastic susceptibility Eq.(5.14) in orders of  $\hat{V}(\mathbf{e})$ . Up to the first order in  $\hat{V}(\mathbf{e})$  we write these expansions in terms of unit block susceptibilities. The



recursion relations for unit block and super block susceptibilities are as follows:

$$\begin{aligned}
\chi_{ijkl}^{\text{super}} &= \chi_{ijkl}^{\text{super rel}} + \chi_{ijkl}^{\text{super res}} \\
&= \chi_{ijkl}^{\text{rel}} - \frac{L_n^3}{N_0^3} \left[ - \sum_{mnpq} \sum_{ss'} \Lambda_{mnpq}^{(ss')}(\mathbf{e}) \right] (\chi_{ijmn}^{\text{rel}} \chi_{pqkl}^{\text{rel}} + \chi_{ijmn}^{\text{rel}} \chi_{pqkl}^{\text{res}} + \chi_{ijmn}^{\text{res}} \chi_{pqkl}^{\text{rel}}) \\
&+ \chi_{ijkl}^{\text{res}} - \frac{L_n^3}{N_0^3} \left[ - \sum_{mnpq} \sum_{ss'} \Lambda_{mnpq}^{(ss')}(\mathbf{e}) \right] \chi_{ijmn}^{\text{res}} \chi_{pqkl}^{\text{res}} \\
&+ \frac{1}{(N_0 L_n)^3} \sum_{mnpq} \sum_{ss'} \left\langle \frac{\delta^2 \Lambda_{mnpq}^{(ss')}(\mathbf{e})}{\delta e_{ij} \delta e_{kl}} \hat{T}_{mn}^{(s)} \hat{T}_{pq}^{(s')} \right\rangle
\end{aligned} \tag{5.16}$$

where again we use  $\chi_{ijkl}^{\text{rel}}$ ,  $\chi_{ijkl}^{\text{res}}$ ,  $\chi_{ijkl}^{\text{super}}$  to stand for  $\chi_{ijkl}^{\text{rel}}(\mathbf{e})$ ,  $\chi_{ijkl}^{\text{res}}(\mathbf{e})$ ,  $\chi_{ijkl}^{\text{super}}(\mathbf{e})$ . For details of calculations please refer to Appendix (B). The last term of Eq.(5.16) is renormalization irrelevant. Compared to other terms in Eq.(5.16) the last term decreases cubically  $L^{-3}$  as the increase of sample length scale  $L$ . To prove this result let us provide a qualitative analysis: denote  $\Lambda_{ijkl}^{(ss')} = -\tilde{\Lambda}_{ijkl}(\vec{n})/8\pi\rho c_t^2 R_{ss'}^3$ , where  $R_{ss'} = |\vec{R}_s - \vec{R}'_s|$  and  $\tilde{\Lambda}_{ijkl}(\vec{n})$  is a dimensionless number of order 1. By applying linear response theory on the last term of Eq.(5.16) with respect to perturbation  $\sum_{ij} \sum_s e_{ij}^{(s)} \hat{T}_{ij}^{(s)}$  to calculate the thermal and quantum averages, it turns out to be the convolution of the imaginary part resonance susceptibilities functional of frequency  $\Omega$ :

$$\sum_{mnpq} \int d\Omega \text{Im} \chi_{ijmn}^{\text{res}}(\Omega) \left( \sum_{ss'} \frac{\hbar L_n^3 \lambda_{mnpq}}{8\pi\rho^2 c_t^4 R_{ss'}^6} \right) \text{Im} \chi_{pqkl}^{\text{res}}(-\Omega) \tag{5.17}$$

where  $\lambda_{mnpq}(\vec{n})$  is the second order derivative of  $\tilde{\Lambda}_{mnpq}(\vec{n})$  with respect to phonon strain field, and it is also a dimensionless number of order 1. We use the assumption that the reduced imaginary part resonance susceptibility  $\text{Im} \tilde{\chi}_{ijkl}^{\text{res}}(\omega) = \text{Im} \chi_{ijkl}^{\text{res}}(\omega)/(1 - e^{-\beta\hbar\omega})$  is approximately a constant up to the frequency  $\omega_c \sim 10^{15}\text{Hz}$  and the temperature of order 10K. Since the imaginary part of resonance susceptibility is always smaller than the reduced version:  $\text{Im} \chi_{ijkl}^{\text{res}}(\omega) < \text{Im} \tilde{\chi}_{ijkl}^{\text{res}}(\omega)$  for arbitrary temperature and frequency, integrating over  $\Omega$  gives the upper limit of Eq.(5.19):  $-C\hbar\omega_c (\text{Im} \tilde{\chi}_t^{\text{res}})^2 / \rho^2 c_t^4 L_n^3$ , where  $C$  is also a dimensionless constant of order 1. If we require that there is a critical length scale  $L_c$ , below which the last term of Eq.(5.16) is comparable to the other terms, the order of magnitude for  $L_c$  is

$$L_c < \left( \frac{\hbar\omega_c}{\rho c_{l,t}^2} \right)^{\frac{1}{3}} \approx 4.6\text{\AA} < L_1 = 50\text{\AA} \tag{5.18}$$

which means the upper limit of  $L_c$  is even smaller than the starting effective length scale of renormalization technique. Throughout the entire renormalization procedure the last term in Eq.(5.16) is always negligible. With the above simplifications one can rewrite the non-elastic susceptibility renormalization equation as

follows,

$$\begin{aligned}
\chi_{ijkl}^{\text{super}} &= \chi_{ijkl}^{\text{super rel}} + \chi_{ijkl}^{\text{super res}} \\
&= (\chi_{ijkl}^{\text{rel}} + \chi_{ijkl}^{\text{res}}) - \frac{L_n^3}{N_0^3} \left[ - \sum_{mnpq} \sum_{ss'} \Lambda_{mnpq}^{(ss')}(\mathbf{e}) \right] (\chi_{ijmn}^{\text{rel}} \chi_{pqkl}^{\text{rel}} + \chi_{ijmn}^{\text{rel}} \chi_{pqkl}^{\text{res}} + \chi_{ijmn}^{\text{res}} \chi_{pqkl}^{\text{rel}} + \chi_{ijmn}^{\text{res}} \chi_{pqkl}^{\text{res}}) \\
&= \chi_{ijkl} - \frac{L_n^3}{N_0^3} \left[ - \sum_{mnpq} \sum_{ss'} \Lambda_{mnpq}^{(ss')}(\mathbf{e}) \right] \chi_{ijmn} \chi_{pqkl}
\end{aligned} \tag{5.19}$$

where the zero-frequency non-elastic susceptibility  $\chi_{ijkl} = \chi_{ijkl}^{\text{rel}} + \chi_{ijkl}^{\text{res}}$ .

The renormalization equation for non-elastic susceptibility can be simplified with the following three steps. First of all, we define a 4-indices tensor  $M_{mnpq}$ , given by

$$M_{mnpq} = \frac{L_n^3}{N_0^3} \left[ - \sum_{ss'} \Lambda_{mnpq}^{(ss')}(\mathbf{e}) \right] \tag{5.20}$$

So the non-elastic susceptibility renormalization relation is rewritten as

$$\chi_{ijkl}^{\text{super}} = \chi_{ijkl} - \chi_{ijmn} M_{mnpq} \chi_{pqkl} \tag{5.21}$$

Second, we denote the 2-fold indices  $(ij), (kl), (mn), (pq)$  in Eq.(5.21) to be  $(ij) \rightarrow A, (kl) \rightarrow B, (mn) \rightarrow C, (pq) \rightarrow D$ . With this simplification, we rewrite 4-indices quantities  $\chi_{ijkl}$  and  $M_{mnpq}$  into a 2-indices matrix form:  $\chi_{AB}$  and  $M_{CD}$ . They are  $6 \times 6$  matrices, for example,  $M_{CD}$  has the indices  $C$  (or  $D$ ) =  $(xx), (xy), (xz), (yy), (yz), (zz)$ . Third, let us define the change of non-elastic susceptibility  $\delta\chi = \chi^{\text{super}} - \chi$ . The real space renormalization equation for non-elastic susceptibility is simplified as:

$$\begin{aligned}
\chi^{\text{super}} = \chi - \chi \mathbf{M} \chi &\Rightarrow \delta\chi = -\chi \mathbf{M} \chi \Rightarrow (\chi)^{-1} \delta\chi (\chi)^{-1} = -\mathbf{M} \Rightarrow \delta(\chi^{-1}) = \mathbf{M} \\
&\Rightarrow \chi^{-1}(R) = \mathbf{M} \log_{N_0} \left( \frac{R}{L_1} \right) + \chi'^{-1}
\end{aligned} \tag{5.22}$$

where the experimental length scale  $R$  is the size of glass sample. In this chapter we consider the avalanche problem with the presence of external static, uniform strain, which means the ‘‘effective phonon wavelength of external strain’’ is much, much greater than the actual size of experimental sample. Therefore, to calculate the matrix  $\mathbf{M}$  we first take the momentum  $k \rightarrow 0$  limit, then take the spacial integral over the non-elastic stress-stress interaction coefficient  $\Lambda_{ijkl}^{(ss')}$ . Please note that the above renormalization irrelevant behavior is only valid for negative eigenvalues of matrix  $\mathbf{M}$  and its corresponding eigenvectors. For the positive eigenvalues of matrix  $\mathbf{M}$ , the renormalization equation, Eq.(5.22) turns out to be problematic: since the

zero-frequency relaxation and resonance susceptibilities are negative, the positive eigenvalues of  $\mathbf{M}$  will lead to the “more negative behavior” to the eigenvalues of non-elastic susceptibility  $\chi$ . Eventually at the experimental length scale  $R$  these negative eigenvalues of  $\chi$  are so large, that the physics picture turns out to be problematic: as long as an infinitesimal strain is applied, the glass system crashes instantly. Here we would like to argue, that for those positive eigenvalues of matrix  $\mathbf{M}$ , our generic couple block model and the renormalization equation may not be applicable in avalanche problem. For the negative eigenvalues of  $\mathbf{M}$  which lead to the logarithmic decreasing behavior of the eigenvalues of non-elastic susceptibility, we try to continue our work to obtain the positive-negative transitions in non-elastic susceptibility.

We have no idea what the value of the constant of integration  $\chi'$  is. One may guess, that the this constant of integration  $\chi'$  is something positive quantity. But this cannot be true. Because the non-elastic susceptibility keeps negative throughout the entire process of renormalization procedure. At experimental length scale, it must be negative as well. It is impossible to find any positive-negative transition in non-elastic susceptibility. If, we use the wrong assumption, that the constant of integration  $\chi'$  is some positive quantity, and it takes the generic isotropic form  $\chi'_{ijkl} = (\chi'_l - 2\chi'_t)\delta_{ij}\delta_{kl} + \chi'_t(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$ , then the non-elastic susceptibility obtained from the wrong assumption is then given by Eq.(5.22).

## 5.5 The Critical External Strain of Avalanche

If we want to prove the existence of positive-negative transition in glass total stress-stress susceptibility, we are actually required to find the singularity in glass total susceptibility. Since the elastic susceptibility  $\chi'_{ijkl} = -(\rho c_l^2 - 2\rho c_t^2)\delta_{ij}\delta_{kl} - \rho c_t^2(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$  does not show such kind of singularity, our hope is to find the singularity in full non-elastic stress-stress susceptibility. However, as we have discussed earlier, the non-elastic susceptibility keeps negative. It is impossible to find singularities in non-elastic susceptibility as well. Let us pretend that in Eq.(5.22) the constant of integration  $\chi'$  is a positive quantity. We will be able to observe the positive-negative transitions in non-elastic susceptibility as follows. But the reader should keep in mind that the results in this section are not correct.

The spherical glass is deformed by external static strain  $e_{xx}$  to become an ellipsoid. Take continuum limit in Eq.(5.20) and change the variables  $\vec{r}_s + \vec{r}'_s = \vec{R}$  and  $\vec{r}_s - \vec{r}'_s = \vec{r}$ , we calculate the matrix  $M_{mnpq}$  as follows

$$M_{mnpq} = \frac{1}{2\pi\rho c_t^2} \int_{V(\mathbf{e})} d^3r \frac{\tilde{\Lambda}_{mnpq}(\vec{r})}{r^3} \quad (5.23)$$

where the integral domain  $V(\mathbf{e})$  is an ellipsoid, for the form of  $\Lambda_{ijkl}^{(ss')}$  please refer to Eq.(3.12, 3.13).

$M \log_{N_0}(R/L_1)$  is then represented by the following matrix form

$$M \log_{N_0} \left( \frac{R}{L_1} \right) = \frac{2}{\rho c_t^2} \ln \left( \frac{R}{L_1} \right) \begin{pmatrix} A & 0 & 0 & B & 0 & B \\ 0 & C & 0 & 0 & 0 & 0 \\ 0 & 0 & C & 0 & 0 & 0 \\ B & 0 & 0 & D & 0 & E \\ 0 & 0 & 0 & 0 & F & 0 \\ B & 0 & 0 & E & 0 & D \end{pmatrix} \quad (5.24)$$

where

$$\begin{aligned} A &= 1 - 3\bar{n}_x^2 + \frac{1}{2}\alpha \left( -3 + 18\bar{n}_x^2 - 15\bar{n}_x^4 \right) \\ B &= \frac{1}{2}\alpha \left[ -1 - 15\bar{n}_x^2\bar{n}_y^2 + 3 \left( \bar{n}_x^2 + \bar{n}_y^2 \right) \right] \\ C &= \frac{1}{4} \left( 2 - 3\bar{n}_x^2 - 3\bar{n}_y^2 \right) + \frac{1}{2}\alpha \left[ -1 - 15\bar{n}_x^2\bar{n}_y^2 + 3 \left( \bar{n}_x^2 + \bar{n}_y^2 \right) \right] \\ D &= 1 - 3\bar{n}_y^2 + \frac{1}{2}\alpha \left( -3 + 18\bar{n}_y^2 - 15\bar{n}_y^4 \right) \\ E &= \frac{1}{2}\alpha \left( -1 - 15\bar{n}_y^2\bar{n}_z^2 + 6\bar{n}_y^2 \right) \\ F &= \frac{1}{2} \left( 1 - 3\bar{n}_y^2 \right) + \frac{1}{2}\alpha \left( -1 - 15\bar{n}_y^2\bar{n}_z^2 + 6\bar{n}_y^2 \right) \end{aligned} \quad (5.25)$$

In the above result we have applied rotational invariance of the integral domain  $V(\mathbf{e})$  around  $x$ -axis, and the parameter  $\alpha = 1 - c_t^2/c_l^2$ . The definition of average values  $\bar{n}_{x,y}^2$ ,  $\bar{n}_{x,y}^4$ ,  $\bar{n}_x^2\bar{n}_y^2$ ,  $\bar{n}_y^2\bar{n}_z^2$  are given as follows: for arbitrary function  $f(\vec{r})$ , the average value is

$$\bar{f}(\vec{r}) = \frac{\int_{V(\mathbf{e})} d^3r f(\vec{r})/r^3}{\int_{V(\mathbf{e})} d^3r 1/r^3} \quad (5.26)$$

Taking integrals over the ellipsoid space, the unit vector averages are displayed as follows,

$$\begin{aligned} \bar{n}_x^2 &= \frac{\epsilon\sqrt{1-\epsilon^2}(-1+2\epsilon^2) + \arcsin \epsilon}{4\epsilon^2 (\epsilon\sqrt{1-\epsilon^2} + \arcsin \epsilon)} \\ \bar{n}_x^4 &= \frac{\epsilon\sqrt{1-\epsilon^2}(-3-2\epsilon^2+8\epsilon^4) + 3\arcsin \epsilon}{24\epsilon^4 (\epsilon\sqrt{1-\epsilon^2} + \arcsin \epsilon)} \\ \bar{n}_y^2\bar{n}_z^2 &= \frac{\epsilon\sqrt{1-\epsilon^2}(-3+10\epsilon^2+8\epsilon^4)}{192\epsilon^4 (\epsilon\sqrt{1-\epsilon^2} + \arcsin \epsilon)} + \frac{3(1-4\epsilon^2+8\epsilon^4)\arcsin \epsilon}{192\epsilon^4 (\epsilon\sqrt{1-\epsilon^2} + \arcsin \epsilon)} \end{aligned} \quad (5.27)$$

where  $0 \leq \epsilon \leq 1$  is the eccentricity of the ellipsoid  $xy$  and  $xz$  cross section (see Fig.5.2). The matrix form of

the inverse of unknown susceptibility  $\chi'$  is given as follows, where we denote  $\alpha' = 1 - \chi'_t/\chi'_l$ ,

$$(\chi^{\text{el}})^{-1} = \frac{1}{\chi'_t} \begin{pmatrix} \frac{\alpha'}{4\alpha'-1} & 0 & 0 & -\frac{2\alpha'-1}{2(4\alpha'-1)} & 0 & -\frac{2\alpha'-1}{2(4\alpha'-1)} \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ -\frac{2\alpha'-1}{2(4\alpha'-1)} & 0 & 0 & \frac{\alpha'}{4\alpha'-1} & 0 & -\frac{2\alpha'-1}{2(4\alpha'-1)} \\ 0 & 0 & 0 & 0 & 1 & 0 \\ -\frac{2\alpha'-1}{2(4\alpha'-1)} & 0 & 0 & -\frac{2\alpha'-1}{2(4\alpha'-1)} & 0 & \frac{\alpha'}{4\alpha'-1} \end{pmatrix} \quad (5.28)$$

where we don't know the exact values of  $\chi'_{l,t}$ . In fact, the non-elastic susceptibility must keep negative. So the assumption that  $\chi'$  is a positive quantity is not correct. The inverse of full non-elastic stress-stress susceptibility  $\chi^{-1}$  is the summation of Eq.(5.24) and Eq.(5.28). For an arbitrary invertible matrix  $\mathbf{A}$ , if  $\varphi$  is one of the eigenvectors of  $\mathbf{A}$ , and  $\lambda$  is the corresponding eigenvalue, then  $\mathbf{A}\varphi = \lambda\varphi$ . We have the important following relation

$$\mathbf{A}^{-1}\varphi = \frac{1}{\lambda}\mathbf{A}^{-1}\lambda\varphi = \frac{1}{\lambda}\mathbf{A}^{-1}\mathbf{A}\varphi = \frac{1}{\lambda}\varphi \quad (5.29)$$

which means as long as  $\varphi$  is the eigenvector of invertible matrix  $\mathbf{A}$ , it is also the eigenvector of  $\mathbf{A}^{-1}$ , with the eigenvalue  $\lambda^{-1}$ . Our purpose is to find the singularity of eigenvalues of full non-elastic susceptibility. From the above proof, if we are able to find the zero-point of eigenvalues of the inverse of full non-elastic susceptibility, which is the summation of Eq.(5.24) and Eq.(5.28), then we can prove the existence of position-negative transition in full non-elastic susceptibility. In the following we straightforwardly calculate the eigenvalues of full non-elastic susceptibility, instead of calculating the eigenvalues of inverse full non-elastic susceptibility.

If we expand Eq.(5.27) in orders of eccentricity and keep only up to the second order of  $\epsilon$ , we will not be able to obtain the singularities of the eigenvalues of full non-elastic susceptibility. Since mechanical avalanche happens when the stress-stress susceptibility of material presents a positive-negative transition at certain critical external strain field  $\mathbf{e}_{\text{crit}}$ , we need to figure out which of the eigenvalues of full non-elastic susceptibility  $\chi$  show such transitions. Among 6 eigenvalues of full non-elastic susceptibility, 3 of them keep positive for eccentricity varies from 0 to 1, while other 3 show positive-negative transitions. We first list a series of variable changes for convenience:  $A' = A + \frac{1}{2\ln(R/L_1)} \frac{\rho c_t^2}{\chi'_t} \frac{\alpha'}{4\alpha'-1}$ ,  $B' = B - \frac{1}{2\ln(R/L_1)} \frac{\rho c_t^2}{\chi'_t} \frac{2\alpha'-1}{2(4\alpha'-1)}$ ,  $C' = C + \frac{1}{2\ln(R/L_1)} \frac{\rho c_t^2}{\chi'_t}$ ,  $D' = D + \frac{1}{2\ln(R/L_1)} \frac{\rho c_t^2}{\chi'_t} \frac{\alpha'}{4\alpha'-1}$ ,  $E' = E - \frac{1}{2\ln(R/L_1)} \frac{\rho c_t^2}{\chi'_t} \frac{2\alpha'-1}{2(4\alpha'-1)}$ ,  $F' = F + \frac{1}{2\ln(R/L_1)} \frac{\rho c_t^2}{\chi'_t}$ , and  $\Delta = 8B'^2 + (A' - D' - E')^2$ . The 6 eigenvalues and corresponding eigenvectors of the matrix form of full non-elastic susceptibility (not the inverse of full non-elastic susceptibility) are given as follows:

eigenvalue	eigenvector
$C'^{-1}$	$(0, 0, 1, 0, 0, 0)$
$C'^{-1}$	$(0, 1, 0, 0, 0, 0)$
$\left(\frac{A'+D'+E'+\sqrt{\Delta}}{2}\right)^{-1}$	$\left(\frac{A'-D'-E'+\sqrt{\Delta}}{2B'}, 0, 0, 1, 0, 1\right)$
$\left(\frac{A'+D'+E'-\sqrt{\Delta}}{2}\right)^{-1}$	$\left(\frac{A'-D'-E'-\sqrt{\Delta}}{2B'}, 0, 0, 1, 0, 1\right)$
$(D' - E')^{-1}$	$(0, 0, 0, -1, 0, 1)$
$F'^{-1}$	$(0, 0, 0, 0, 1, 0)$

(5.30)

As an example, we choose the average value of  $\alpha = 1 - c_t^2/c_l^2 = 0.7$  and  $R = 1\text{mm}$  so  $\ln(R/L_1) \approx 12$  for amorphous solids. We also choose, for example,  $\chi'_t = \rho c_t^2$  and  $\chi'_l = \rho c_l^2$  to give an illustration of the following positive-negative transitions of non-elastic susceptibility. The first, second and third eigenvalues  $C'^{-1}$ ,  $C'^{-1}$  and  $\left(\frac{A'+D'+E'+\sqrt{\Delta}}{2}\right)^{-1}$  stay positive for eccentricity varies from 0 to 1. The plots of eigenvalue versus eccentricity are displayed as follows.

Fig.5.3 and Fig.5.4 are positive eigenvalues of matrix  $\mathbf{M}$ . The corresponding strain directions are  $e_{xy}$ ,  $e_{xz}$  and  $\frac{A'-D'-E'+\sqrt{\Delta}}{2B'}e_{xx} + e_{yy} + e_{zz}$ , where the coefficient  $\frac{A'-D'-E'+\sqrt{\Delta}}{2B'} < 0$  for  $\forall \epsilon \in [0, 1]$ . We plot the negativity of coefficient  $\frac{A'-D'-E'+\sqrt{\Delta}}{2B'}$  in Fig. 5.4,

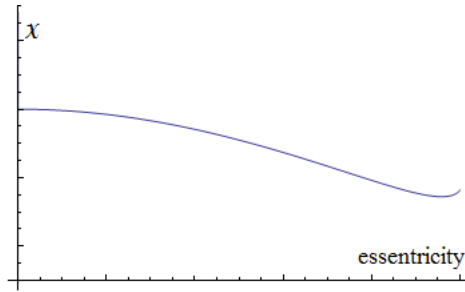


Figure 5.3: The first and second eigenvalues  $C'^{-1}$  in units of  $\rho c_t^2$  as the function of eccentricity ( $x$ -axis) varies from 0 to 1. It stays positive.

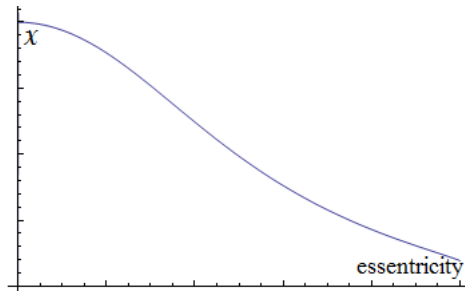


Figure 5.4: The third eigenvalue  $\left(\frac{A'+D'+E'+\sqrt{\Delta}}{2}\right)^{-1}$  as the function of eccentricity. It stays positive.

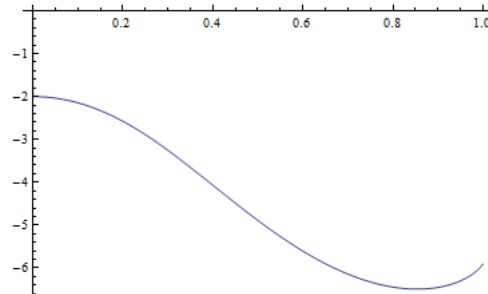


Figure 5.5: The **coefficient** in the third eigenvector,  $\frac{A'-D'-E'+\sqrt{\Delta}}{2B'}$  as the function of eccentricity. It stays negative for eccentricity  $\forall \epsilon \in [0, 1]$ , with the value from  $-2$  to  $-6$ .

On the other hand, the fourth, fifth and sixth eigenvalues of matrix  $\mathbf{M}$  are negative. The corresponding eigenvalues  $\left(\frac{A'+D'+E'-\sqrt{\Delta}}{2}\right)^{-1}$ ,  $(D' - E')^{-1}$  and  $F'^{-1}$  of non-elastic susceptibility  $\chi$  present positive-negative transitions at certain critical eccentricity varies from 0 to 1:

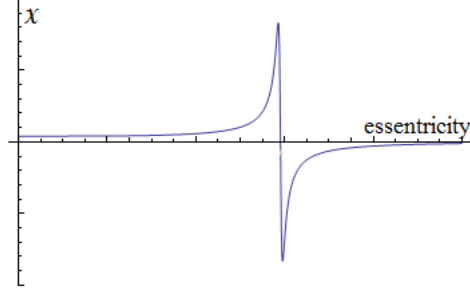


Figure 5.6: The fourth eigenvalue  $\left(\frac{A'+D'+E'-\sqrt{\Delta}}{2}\right)^{-1}$  as the function of eccentricity.

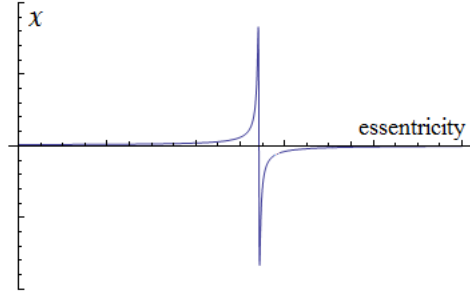


Figure 5.7: The fifth eigenvalue  $(D' - E')^{-1}$  as the function of eccentricity..

Let's discuss the eigenvalues which show positive-negative transitions in details. First, the eigenvector which corresponds to the eigenvalue  $\left(\frac{A'+D'+E'-\sqrt{\Delta}}{2}\right)^{-1}$  is  $\left(\frac{A'-D'-E'-\sqrt{\Delta}}{2B'}, 0, 0, 1, 0, 1\right)$ . From Fig. 5.9, the coefficients of  $e_{xx}$  and  $e_{yy}$  and  $e_{zz}$  have the same signs. For the external static strain which pulls glass system in  $x$  direction with  $e_{xx}$ , when it exceeds critical value, the glass is fragile against additional expansion or contraction deformations. Second, the eigenvector which corresponds to the eigenvalue  $(D' - E')^{-1}$  is  $(0, 0, 0, -1, 0, 1)$ . When the external static strain exceeds critical value, the glass is fragile against additional external strain  $\pm(e_{yy} - e_{zz})$ , which is to pull glass in  $y$  or  $z$  direction and squeeze in another direction. Third, the eigenvector for eigenvalue  $F'^{-1}$  is  $e_{yz}$ , a shear deformation to glass system. For the external static strain exceeding critical value, the glass is fragile against additional shear in  $yz$  plane.



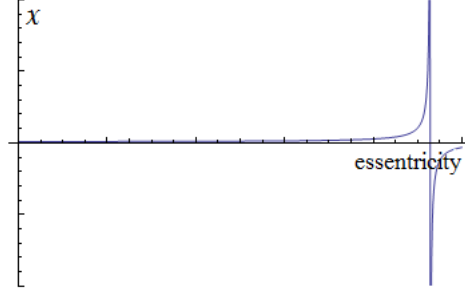


Figure 5.8: The sixth eigenvalue  $F'^{-1}$  as the function of eccentricity.

Fig.5.6-5.8 indicate when external static deformation  $e_{xx} = e$  exceeds certain critical value, glass is fragile against the external strain fields in the directions of  $\frac{A'-D'-E'-\sqrt{\Delta}}{2B'}e_{xx} + e_{yy} + e_{zz}$ ,  $-e_{yy} + e_{zz}$  and  $e_{yz}$ .

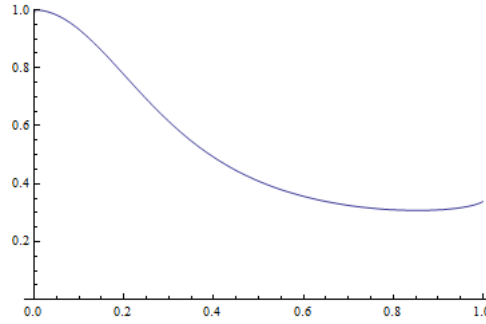


Figure 5.9: The coefficient in the sixth eigenvector,  $\frac{A'-D'-E'-\sqrt{\Delta}}{2B'}$  as the function of  $\epsilon$ . It stays positive for eccentricity  $\forall \epsilon \in [0, 1]$ .

Finally, to verify the existence of mechanical avalanche phenomena, we need to sum up elastic and non-elastic susceptibilities to get total susceptibility,  $\chi^{\text{tot}} = \chi^{\text{el}} + \chi$ . However the elastic susceptibility does not have a singularity. For external strain fields away from the critical value, there is no steep positive-negative transition in glass mechanical susceptibility. When external strain approaches critical value, non-elastic susceptibility presents a sharp positive-negative transition. The singularity of total susceptibility (glass avalanche behavior) is therefore determined by the singularity of non-elastic susceptibility. (However, please note this result is based on the wrong assumption that the constant of integration  $\chi'$  is a positive quantity in the renormalization Eq.(5.22). The motivation of making this assumption that  $\chi'$  is positive is because we want to obtain the positive-negative transition in non-elastic susceptibility. We hope to get some useful results to explain the glass mechanical avalanche problem, but since the non-elastic susceptibility keeps negative from the starting microscopic length scale, the assumption that  $\chi'$  is positive is not valid. It is at this point that finally our theory is not able to explain the avalanche problem. )

This chapter is only a tentative work to apply our generic coupled block model into a new field, glass

avalanche problem. Our model is only valid below the temperature of  $T \approx 50\text{K}$ . However, the glass avalanche experiments we are able to find are taken under room temperatures or glass transition temperatures [21, 43, 44, 39, 40] ( $T \sim 300\text{K}$ ). This might be another reason that our model is not applicable in the glass avalanche problem.

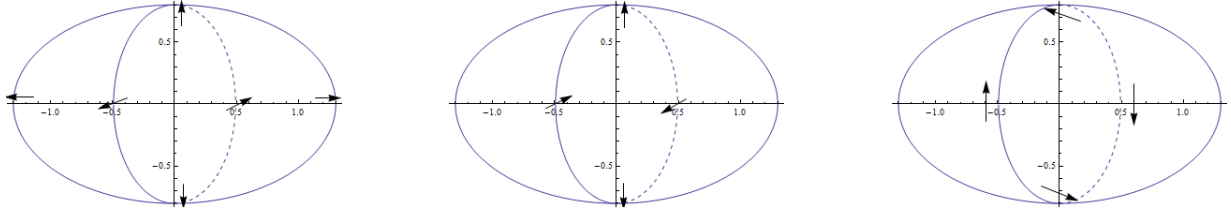


Figure 5.10: Three external strain field directions to crack the glass. (1) pull or squeeze it in  $e_{xx}$ ,  $e_{yy}$  and  $e_{zz}$  strain; (2) pull in  $e_{yy}$  strain direction while squeeze in  $e_{zz}$  direction, or vice versa; (3) shear in  $yz$  plane, please note  $\partial u_y/\partial z$  and  $\partial u_z/\partial y$  not necessarily the same.

## Chapter 6

# Universal Meissner-Berret Ratio

It has been more than 50 years since the first experiment[1] by Zeller and Pohl showed at ultra-low temperatures below 1K the thermal properties of amorphous solids behave entirely different from that of crystalline counterparts. Anderson, Halperin and Varma[3] group and Phillips[19] independently developed a model which was later known as tunneling-two-level-system (TTLS) model. It successfully explained several universal experimental results of amorphous solids which cannot be found in crystalline solids, e.g., linear heat capacity, saturation, echoes, low-temperature heat conductivity etc. In TTLS model people assume the Hamiltonian of amorphous solid is the summation of elastic (phonon) part of Hamiltonian, a set of non-elastic two-level-systems and phonon-TTLS couplings. The longitudinal and transverse phonon-TTLS coupling constants  $\gamma_{l,t}$  are adjustable parameters. However, in 1987 it was Meissner and Berret's experiment[45] first pointed out the coupling constants  $\gamma_{l,t}$  are not arbitrary: below temperature  $T < 1K$ , the ratio between them  $\gamma_l/\gamma_t$  turns out to lie between 1.44 and 1.84 for a wide variety of amorphous materials, regardless of their chemical compounds and microscopic molecular structure. Such universality suggests coupling constants  $\gamma_{l,t}$  come from more general mechanism which cannot be explained within TTLS model. In the rest of this chapter, we use "Meissner-Berret Ratio" to represent for "the ratio  $\gamma_l/\gamma_t$  of TTLS coupling constant".

We want to investigate the universality of Meissner-Berret ratio ( $\gamma_l/\gamma_t \approx (1.44 \sim 1.84)$ ) by applying our generic coupled block model. Within TTLS model the resonance energy absorption per unit time  $\dot{E}_{l,t}$  is proportional to the square of coupling constant  $\gamma_{l,t}$ ; in our model this energy absorption rate is proportional to the imaginary part of non-elastic resonance susceptibility  $\text{Im } \chi_{l,t}^{\text{res}}$ , which will be defined in details in section 1. So if we want to prove the "universality of  $\gamma_l/\gamma_t$ ", we are actually proving the "universality of  $\text{Im } \chi_l^{\text{res}}/\text{Im } \chi_t^{\text{res}}$ " in our generic coupled block model, where  $\chi_l$  is the non-elastic compression modulus, and  $\chi_t$  is non-elastic shear modulus.

## 6.1 The Set up of Meissner-Berret Ratio Problem

Based on TTLS model the amorphous solid (glass) Hamiltonian with the coupling between two-level-system and phonon strain field is written as[10]

$$\hat{H} = \frac{1}{2} \begin{pmatrix} E & 0 \\ 0 & -E \end{pmatrix} + \frac{\gamma_{l,t}}{2} Ak \begin{pmatrix} D & M \\ M & -D \end{pmatrix} e^{i\omega t} \quad (6.1)$$

where the Hamiltonian is written in two-level-system energy eigenbasis with  $E = \sqrt{\Delta^2 + \Delta_0^2}$ ;  $D = \Delta/E$  and  $M = \Delta_0/E$  are diagonal and off-diagonal matrix elements[20] of coupling between two-level-system and phonon strain field, and by definition they are no greater than 1;  $Ak$  is the product of phonon wave amplitude  $A$  and wave number  $k$ ;  $\omega$  is the frequency of input external phonon;  $\gamma_{l,t}$  is the coupling constants for longitudinal/transverse phonon strains. Because in glass there are a set of TTLS with different parameters  $\Delta, \Delta_0, \gamma_{l,t}$ , those TTLS in resonance with external phonon field  $E = \hbar\omega$  can resonantly absorb phonon energy which linearly increases with time  $t$ . Using Fermi golden rule the resonance energy absorption per unit time is proportional to coupling constant squared:

$$\dot{E}_{l,t} = \frac{\pi}{2\hbar} A^2 k^2 M^2 E \tanh\left(\frac{1}{2}\beta\hbar\omega\right) \delta(E - \hbar\omega) \gamma_{l,t}^2 \propto \gamma_{l,t}^2 \quad (6.2)$$

where we take phonon strain  $e = Ak$  and frequency  $\omega$  to be identical for longitudinal and transverse input phonons.

Since the set up of TTLS model is based on these parameters, within it we cannot explain the universality of  $\gamma_l/\gamma_t$ . Therefore, we want to apply our generic coupled block model to consider their energy absorption due to external phonon fields, and try to explore if the ratio of energy absorption due to longitudinal and transverse phonon turns out to be universal or material independent.

Let us consider a block of glass with the dimension  $L$  much greater than the atomic distance  $a \sim 10\text{\AA}$ . The elastic strain  $e_{ij}(\vec{x}, t)$  can be defined as the spacial derivative of displacement  $\vec{u}(\vec{x}, t)$  for the matter located at  $\vec{x}$ :  $e_{ij}(\vec{x}, t) = \frac{1}{2} \left( \frac{\partial u_i(\vec{x}, t)}{\partial x_j} + \frac{\partial u_j(\vec{x}, t)}{\partial x_i} \right)$ . We write  $\hat{H}^{\text{tot}}$  for the total Hamiltonian of the glass block, and expand it in orders of elastic intrinsic strain field  $e_{ij}(\vec{x}, t)$  in long wavelength limit:

$$\hat{H}^{\text{tot}} = \hat{H}_0^{\text{tot}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{tot}}(\vec{x}) + \mathcal{O}(e_{ij}^2) \quad (6.3)$$

where the definition of stress tensor  $\hat{T}_{ij}^{\text{tot}}(\vec{x})$  is the first order derivative of Hamiltonian with respect to

intrinsic strain field

$$\hat{T}_{ij}^{\text{tot}}(\vec{x}) = \frac{\delta \hat{H}^{\text{tot}}}{\delta e_{ij}(\vec{x})} \quad (6.4)$$

Next we can define stress-stress susceptibility, the derivative of stress tensor  $\hat{T}_{ij}^{\text{tot}}$  with respect to intrinsic strain field  $e_{kl}$ . The susceptibility is taken for the glass block much larger than atomic distance:

$$\chi_{ijkl}^{\text{tot}}(\vec{x} - \vec{x}'; t - t') = \frac{\delta \langle \hat{T}_{ij}^{\text{tot}} \rangle(\vec{x}, t)}{\delta e_{kl}(\vec{x}', t')} \quad (6.5)$$

In the above definition the average operator  $\langle \rangle$  represents thermal average and quantum average. For an arbitrary operator  $\hat{A}$ ,  $\langle \hat{A} \rangle = \sum_m \mathcal{Z}^{-1} e^{-\beta E_m} \langle m, t | \hat{A} | m, t \rangle$  with  $|m\rangle$  the eigenbasis of Hamiltonian  $\hat{H}_0^{\text{tot}}$  and  $\mathcal{Z}$  the partition function with the temperature  $\beta = (k_B T)^{-1}$ .

Let us separate Hamiltonian  $\hat{H}^{\text{tot}}$  into purely elastic part  $\hat{H}^{\text{el}}$  and non-elastic part  $\hat{H}^{\text{non}}$ . We define a new stress tensor which comes from non-elastic part of glass Hamiltonian:

$$\begin{aligned} \hat{H}^{\text{non}} &= \hat{H}_0^{\text{non}} + \int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}^{\text{non}}(\vec{x}) + \mathcal{O}(e_{ij}^2) \\ \hat{T}_{ij}^{\text{non}}(\vec{x}) &= \frac{\delta \hat{H}^{\text{non}}}{\delta e_{ij}(\vec{x})} \end{aligned} \quad (6.6)$$

The non-elastic stress-stress susceptibility is then defined as  $\chi_{ijkl}^{\text{non}}(\vec{x} - \vec{x}'; t - t') = \delta \langle \hat{T}_{ij}^{\text{non}} \rangle(\vec{x}, t) / \delta e_{kl}(\vec{x}', t')$ . In the rest of this chapter we will always use  $\hat{H}_0$ ,  $\chi_{ijkl}$  and  $\hat{T}_{ij}$  to stand for non-elastic part of  $\hat{H}_0^{\text{non}}$ ,  $\chi_{ijkl}^{\text{non}}$  and  $\hat{T}_{ij}^{\text{non}}$ .

To calculate the space-averaged non-elastic stress-stress susceptibility  $\chi_{ijkl}(\omega) = \frac{1}{L^3} \int d^3x d^3x' \chi_{ijkl}(\vec{x} - \vec{x}'; \omega)$  let's denote  $|m\rangle$  and  $E_m$  to be the eigenbasis and eigenvalues of unperturbed non-elastic Hamiltonian  $\hat{H}_0$ . The space-averaged susceptibility is volume independent. The eigenbasis  $|m\rangle$  is a generic multiple-level-system. By putting in external weak intrinsic strain field  $e_{ij}(\vec{x}, t)$  the system receives a perturbation  $\int d^3x \sum_{ij} e_{ij}(\vec{x}, t) \hat{T}_{ij}(\vec{x})$ . Using linear response theory on  $\langle \hat{T}_{ij} \rangle(\vec{x}, t)$  with respect to perturbation  $e_{ij} \hat{T}_{ij}$ , we obtain (the imaginary part of resonance) non-elastic susceptibility as follows:

$$\begin{aligned} \text{Im} \chi_{ijkl}^{\text{res}}(T, \omega) &= \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \text{Im} \chi_{ijkl}^{(m)}(\omega) \\ \text{Im} \chi_{ijkl}^{(m)}(\omega) &= \frac{\pi}{L^3} \int d^3x d^3x' \sum_n \langle m | \hat{T}_{ij}(\vec{x}) | n \rangle \langle n | \hat{T}_{kl}(\vec{x}') | m \rangle \\ &\quad [-\delta(E_n - E_m - \omega) + \delta(E_n - E_m + \omega)] \end{aligned} \quad (6.7)$$

Please note, that the above definition of imaginary susceptibility is self-consistent with (1) the definition of susceptibility in “Theory of Quantum Liquids” by David Pines and Phillippe Nozieres[61] and (2) the definition of non-elastic susceptibility in chapters 4 and 5 in this thesis. For  $\omega > 0$ , the imaginary part of non-elastic susceptibility is negative-definite (in the book by David Pines, the imaginary susceptibility is negative-definite as well).  $\mathcal{Z} = \sum_n e^{-\beta E_n}$  is the partition function of unperturbed non-elastic Hamiltonian  $\hat{H}_0$ , and we set  $\hbar = 1$ . Because for arbitrary quantum number  $n$  we always have  $E_n \geq E_0$ , the definition of  $\text{Im} \chi_{ijkl}^{(m)}(\omega)$  in Eq.(6.7) is only valid when  $E_m \geq \omega \geq -E_m$ ; when  $E_m < \omega$  or  $-E_m > \omega$ , in the above definition of imaginary part of resonance susceptibility, one of the delta-functions will vanish. Therefore when  $E_m < \omega$  or  $-E_m > \omega$ , the imaginary part of resonance susceptibility is simplified as follows,

$$\begin{aligned} \text{Im} \chi_{ijkl}^{(m)}(\omega) &= \frac{\pi}{L^3} \int d^3x d^3x' \sum_n \langle m | \hat{T}_{ij}(\vec{x}) | n \rangle \langle n | \hat{T}_{kl}(\vec{x}') | m \rangle [+ \delta(E_n - E_m + \omega)] \quad \text{if } \omega < -E_m \\ \text{Im} \chi_{ijkl}^{(m)}(\omega) &= \frac{\pi}{L^3} \int d^3x d^3x' \sum_n \langle m | \hat{T}_{ij}(\vec{x}) | n \rangle \langle n | \hat{T}_{kl}(\vec{x}') | m \rangle [- \delta(E_n - E_m - \omega)] \quad \text{if } \omega > E_m \end{aligned} \quad (6.8)$$

it is convenient to rewrite the imaginary resonance susceptibility Eq.(6.7) into reduced imaginary susceptibility  $\text{Im} \tilde{\chi}_{ijkl}$  as follows for future use:

$$\begin{aligned} \text{Im} \chi_{ijkl}^{\text{res}}(T, \omega) &= (1 - e^{-\beta \hbar \omega}) \text{Im} \tilde{\chi}_{ijkl}(T, \omega) \\ \text{Im} \tilde{\chi}_{ijkl}^{\text{res}}(T, \omega) &= \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \text{Im} \tilde{\chi}_{ijkl}^{(m)}(\omega) \\ \text{Im} \tilde{\chi}_{ijkl}^{(m)}(\omega) &= \frac{\pi}{L^3} \int d^3x d^3x' \sum_n \langle m | \hat{T}_{ij}(\vec{x}) | n \rangle \langle n | \hat{T}_{kl}(\vec{x}') | m \rangle [- \delta(E_n - E_m - \omega)] \end{aligned} \quad (6.9)$$

Please note, that by definition  $\text{Im} \tilde{\chi}_{ijkl}^{\text{res}}(T, \omega)$  is also a negative-definite quantity. Again, for an arbitrary isotropic system the reduced non-elastic susceptibility must satisfy the genetic form

$$\text{Im} \tilde{\chi}_{ijkl}^{\text{res}}(T, \omega) = (\text{Im} \tilde{\chi}_l^{\text{res}}(T, \omega) - 2 \text{Im} \tilde{\chi}_t^{\text{res}}(T, \omega)) \delta_{ij} \delta_{kl} + \text{Im} \tilde{\chi}_t^{\text{res}}(T, \omega) (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) \quad (6.10)$$

According to the negative-definite property of  $\text{Im} \tilde{\chi}_{ijkl}^{\text{res}}(T, \omega)$ , the newly-defined quantities  $\text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega)$  are negative-definite as well. Please note we use  $\text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega)$  to stand for imaginary part of reduced non-elastic longitudinal/transverse susceptibility  $\text{Im} \tilde{\chi}_{l,t}^{\text{res non}}(T, \omega)$ . The real part of reduced non-elastic susceptibility  $\text{Re} \tilde{\chi}_{ijkl}^{\text{res}}(T, \omega)$  can be obtained by Kramers-Kronig relation from the imaginary part of it. Therefore after the Kramers-Kronig transformation, the real part of reduced non-elastic susceptibility  $\text{Re} \tilde{\chi}_{ijkl}^{\text{res}}(T, \omega)$  is also a negative quantity.

## 6.2 Virtual Phonon Exchange Interactions

From the definition Eq.(6.6), within the consideration of single generic block, non-elastic stress tensor  $\hat{T}_{ij}(\vec{x})$  and non-elastic unperturbed Hamiltonian  $\hat{H}_0$  are simply generalizations from 2-level-systems to multiple-level-system (see Eq.(6.1)). Nothing non-trivial will be obtained within single block considerations. However, if we combine a set of such blocks together to form a super block, the interaction between single blocks will be taken into account. Since the stress-strain interacting term  $e_{ij}\hat{T}_{ij}$  contains phonon field  $e_{ij}$ , the exchange of virtual phonons will give an effective RKKY-type interaction between blocks via stress tensor products:

$$\hat{V} = \int d^3x d^3x' \sum_{ijkl} \Lambda_{ijkl}(\vec{x} - \vec{x}') \hat{T}_{ij}(\vec{x}) \hat{T}_{kl}(\vec{x}') \quad (6.11)$$

where the coefficient  $\Lambda_{ijkl}(\vec{x} - \vec{x}')$  has been carried out in Appendix (A). We call Eq.(6.11) non-elastic stress-stress interaction. In the rest of this thesis we always 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, when  $\vec{x}_s$  denotes the center of the  $s$ -th block, and that  $\int_{V^{(s)}} \hat{T}_{ij}(\vec{x}) d^3x = \hat{T}_{ij}^{(s)}$  is the uniform stress tensor of the  $s$ -th block. Also, from now on we use  $e_{ij}^{(s)}(t)$  to denote the phonon strain field  $e_{ij}(\vec{x}, t)$  located at the  $s$ -th block. By combining  $N_0 \times N_0 \times N_0$  identical  $L \times L \times L$  unit blocks to form a  $N_0L \times N_0L \times N_0L$  super block, the Hamiltonian without external strain field is written as

$$\hat{H}^{\text{super}} = \sum_s^{N_0^3} \hat{H}_0^{(s)} + \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \quad (6.12)$$

From now on we make the important assumption that these block space-averaged stress tensors  $\hat{T}_{ij}^{(s)}$  are diagonal in spacial coordinates:  $\text{Im} \tilde{\chi}_{ijkl}^{(ss')}(T, \omega) = \frac{1}{L^3} \langle \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \rangle = \text{Im} \tilde{\chi}_{ijkl}(T, \omega) \delta_{ss'}$ .

Next, let us consider glass Hamiltonian with the presence of external strain field  $e_{ij}(\vec{x}, t)$  as a perturbation. Please note that we have defined non-elastic stress tensor and non-elastic stress-stress susceptibility with the help of intrinsic phonon strain field, in this section  $e(\vec{x}, t)$  stands for the external real phonon field. It seems the Hamiltonian Eq.(6.12) simply adds a stress-strain interacting term  $\sum_s \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)}$ . However, more questions arise with the appearance of external strain field.

First of all these non-elastic stress tensors  $\hat{T}_{ij}^{(s)}$  might be modified. A familiar example is that external strain field can modify electric dipole moments by changing relative positions of positive-negative charge pairs (to the leading order of external strain):  $\Delta p_i(t) = \sum_j (\partial u_i(t) / \partial x_j) p_j$  where  $i, j$  are cartesian coordinates, and  $\vec{u}(\vec{x}, t)$  is phonon field. In principle we need to obtain the modification of stress tensors,  $\Delta \hat{T}_{ij}^{(s)}(t)$  to the leading order in  $e_{ij}^{(s)}(t)$  for the resonance energy absorption contribution. However, we only qualitatively

know the expansion of  $\Delta\hat{T}_{ij}^{(s)}(t)$  in orders of external strain  $e = Ak$  is  $\Delta\hat{T}_{ij}^{(s)}(t) \sim e(t)\hat{T}_{ij}^{(s)} + \mathcal{O}(e^2)$ . Within qualitative Taylor series technique we calculate the energy absorption contribution in Eq.(6.21). We will show this energy absorption contribution is renormalization irrelevant at experimental length scale in section 4 via scaling analysis.

There is a second problem arising from external phonon strain field: the relative positions of unit blocks  $\vec{x}_s - \vec{x}_{s'}$  can be changed, resulting in the modification of stress-stress interaction coefficient  $\Lambda_{ijkl}^{(ss')}(\mathbf{e})$ . To the first order expansion in external strain field the modification of  $\Lambda_{ijkl}^{(ss')}$  is

$$\begin{aligned}
\Delta\Lambda_{ijkl}^{(ss')} &= \left( \frac{x_{ss'}}{\Delta x_{ss'}} \Delta\tilde{\Lambda}_{ijkl}^{(ss')} - 3\tilde{\Lambda}_{ijkl}^{(ss')} \cos\theta_{ss'} \right) \frac{\Delta x_{ss'}}{x_{ss'}^4} \\
\Delta\tilde{\Lambda}_{ijkl}^{(ss')} &= \left\{ \frac{3}{4} \left[ 2(n_j n_l \delta_{ik} + n_j n_k \delta_{il} + n_i n_k \delta_{jl} + n_i n_l \delta_{jk}) \cos\theta_{ss'} \right. \right. \\
&\quad \left. \left. - [(m_j n_l + m_l n_j) \delta_{ik} + (m_j n_k + m_k n_j) \delta_{il} + (m_i n_k + m_k n_i) \delta_{jl} + (m_i n_l + m_l n_i)] \delta_{jk} \right] \right. \\
&\quad \left. - 3\alpha \cos\theta_{ss'} \left( n_k n_l \delta_{ij} + n_j n_l \delta_{ik} + n_k n_j \delta_{il} + n_i n_l \delta_{jk} + n_i n_k \delta_{jl} + n_i n_j \delta_{kl} \right) \right. \\
&\quad \left. + \frac{3}{2} \alpha \left[ m_i (n_l \delta_{jk} + n_k \delta_{jl} + n_j \delta_{kl}) + m_j (n_l \delta_{ik} + n_k \delta_{il} + n_i \delta_{kl}) \right. \right. \\
&\quad \left. \left. + m_k (n_l \delta_{ij} + n_i \delta_{jl} + n_j \delta_{il}) + m_l (n_k \delta_{ij} + n_i \delta_{jk} + n_j \delta_{ik}) \right] \right. \\
&\quad \left. - \frac{15}{2} \alpha \left( m_i n_j n_k n_l + m_j n_i n_k n_l + m_k n_i n_j n_l + m_l n_i n_j n_k \right) + 30\alpha n_i n_j n_k n_l \cos\theta_{ss'} \right\} \frac{\Delta x_{ss'}}{x_{ss'}^4} \quad (6.13)
\end{aligned}$$

where  $\alpha = 1 - c_t^2/c_l^2$ ,  $x_{ss'} = |\vec{x}_s - \vec{x}_{s'}|$ ,  $\Delta\vec{x}_s = \vec{u}(\vec{x}_s, t)$ ,  $\Delta x_{ss'} = |\Delta\vec{x}_s - \Delta\vec{x}_{s'}|$ ,  $\cos\theta_{ss'} = (\Delta\vec{x}_{ss'} \cdot \vec{x}_{ss'}) / \Delta x_{ss'} x_{ss'}$  is the angle between  $\Delta\vec{x}_{ss'}$  and  $\vec{x}_{ss'}$ , and  $\vec{m} = \Delta\vec{x}_{ss'} / \Delta x_{ss'}$  is the unit vector of  $\Delta\vec{x}_{ss'}$ . Finally by taking everything into account the total Hamiltonian for super block amorphous solid with the presence of external weak strain field  $\mathbf{e}(\vec{x}, t)$  reads

$$\begin{aligned}
\hat{H}^{\text{super}}(\mathbf{e}) &= \sum_s^{N_0^3} \left( \hat{H}_0^{(s)} + \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)} \right) \\
&\quad + \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \left( \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} + \Delta\Lambda_{ijkl}^{(ss')}(t) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} + 2\Lambda_{ijkl}^{(ss')} \Delta\hat{T}_{ij}^{(s)}(t) \hat{T}_{kl}^{(s')} \right) \quad (6.14)
\end{aligned}$$

where  $e_{ij}^{(s)}(t)$  is real phonon field here.



## 6.3 Second Order Perturbation Theory to Energy Absorption of Super Block

In previous discussions we know within TTLS model the resonance energy absorption per unit time is proportional to coupling constant squared:  $\dot{E}_{l,t} \propto \gamma_{l,t}^2$ . In this section we use generic coupled block model to consider it from longitudinal and transverse phonons. We first consider a single-block amorphous solid with dimension  $L \times L \times L$ , with the unperturbed Hamiltonian  $\hat{H}_0$  and perturbation  $\sum_{ij} e_{ij}(t)\hat{T}_{ij}$ , so the total Hamiltonian is  $\hat{H} = \hat{H}_0 + \sum_{ij} e_{ij}(t)\hat{T}_{ij}$ . We denote  $|n\rangle$  and  $E_n$  to be the  $n$ -th eigenstate and eigenvalue of unperturbed Hamiltonian  $\hat{H}_0$ . Thus the single-block energy absorption rate is  $\dot{E}_{l,t}^{\text{single}} = \frac{\partial}{\partial t} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \left( \langle n_{I,t} | \hat{H}_I(t) | n_{I,t} \rangle - \langle n | \hat{H}_0 | n \rangle \right)$ , where  $|n_{I,t}\rangle = e^{-\frac{i}{\hbar} \int_{-\infty}^t \sum_{ij} e_{ij}(t') \hat{T}_{ij(I)}(t') dt'} |n\rangle$  is the interaction picture wavefunction, and  $\hat{H}_I(t)$  and  $\hat{T}_{ij(I)}(t')$  are interaction picture operators. For an arbitrary interaction picture operator  $\hat{A}_I(t)$  we have  $\hat{A}_I(t) = e^{i\hat{H}_0 t/\hbar} \hat{A}(t) e^{-i\hat{H}_0 t/\hbar}$ . The resonance energy absorption per unit time of single-block is

$$\dot{E}_{l,t}^{\text{single}} = -2L^3 A^2 k^2 \omega (1 - e^{-\beta \hbar \omega}) \text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega) \quad (6.15)$$

For details of calculations, please see Appendix (C). In the above result, according to the negativity of  $\text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega)$ , the single block energy absorption rate is a positive quantity.  $Ak$  is the strength of external strain field  $e_{ij}$ ,  $\omega$  is phonon frequency. With the argument by D. C. Vural and A. J. Leggett[25] and the experiment by R. O. Pohl, X. Liu and E. Thompson[34] we assume that within a certain extent of frequency  $\omega < \omega_c$  below 1K the longitudinal and transverse imaginary susceptibility can be approximately treated as a constant of frequency  $\text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega) \approx \text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T)$ . In section 4, Eq.(5.27) we will discuss the order of magnitude of  $\omega_c$  in details. Given the external phonon field amplitude  $A$  and wave number  $k$  the energy absorption per unit time for single-block amorphous solid is proportional to longitudinal and transverse imaginary susceptibility  $\text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T)$ :  $\dot{E}_l^{\text{single}} / \dot{E}_t^{\text{single}} = \text{Im} \tilde{\chi}_l^{\text{res}}(T) / \text{Im} \tilde{\chi}_t^{\text{res}}(T)$ . Compare this with energy absorption rate from TTLS model, i.e.,  $\dot{E}_l^{\text{single}} / \dot{E}_t^{\text{single}} = \gamma_l^2 / \gamma_t^2$ , we get the relation between imaginary susceptibility and coupling constant  $\text{Im} \tilde{\chi}_l^{\text{res}}(T) / \text{Im} \tilde{\chi}_t^{\text{res}}(T) = \gamma_l^2 / \gamma_t^2$ .

Within single-block considerations one cannot extract more information from generic block model than TTLS. However, the exchange of virtual phonons allows non-elastic stress-stress interaction between blocks. Let's think about a set of  $N_0^3$  identical single blocks with the dimension  $L \times L \times L$  combined together to form a super block  $N_0 L \times N_0 L \times N_0 L$ . The presence of many-block interaction  $\hat{V}$  affects the energy absorption of super block. To explore this problem we follow three steps: (1) turn off stress-stress (many-block)

interaction  $\hat{V}$ . These  $N_0^3$  identical single blocks are non-interacting. Thus the Hamiltonian for super block is the summation of single block Hamiltonians  $\hat{H}_0 = \sum_s \hat{H}_0^{(s)}$ , where  $s$  denotes the  $s$ -th block which runs over  $s = 1, 2, \dots, N_0^3$ . We denote  $|n\rangle = \prod_s |n^{(s)}\rangle$  and  $E_n = \sum_s E_n^{(s)}$  to be the  $n$ -th eigenstate and eigenvalue for Hamiltonian  $\hat{H}_0$ ; (2) turn on non-elastic stress-stress interaction  $\hat{V} = \sum_{ss'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}$  as static perturbation. The eigenstate and eigenvalue change as follows:

$$|n^*\rangle = |n\rangle + \sum_{l \neq n} \frac{\langle l | \hat{V} | n \rangle}{E_n - E_l} |l\rangle + \dots \quad E_n^* = E_n + \langle n | \hat{V} | n \rangle + \sum_{l \neq n} \frac{|\langle l | \hat{V} | n \rangle|^2}{E_n - E_l} + \dots \quad (6.16)$$

where  $|n^*\rangle$  and  $E_n^*$  are the  $n$ -th eigenstate and eigenvalue for  $\hat{H}_0 + \hat{V}$ ; (3) take  $\hat{H}_0 + \hat{V}$  as static Hamiltonian of interaction picture, we turn on time-dependent perturbation

$$\hat{H}'(t) = \sum_s \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)} + \sum_{ss'} \sum_{ijkl} \left( \Delta \Lambda_{ijkl}^{(ss')}(t) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} + 2 \Lambda_{ijkl}^{(ss')} \Delta \hat{T}_{ij}^{(s)}(t) \hat{T}_{kl}^{(s')} \right) \quad (6.17)$$

to calculate the energy absorption rate of super block Hamiltonian  $\hat{H}_0 + \hat{V}$ :

$$\dot{E}_{l,t}^{\text{super}}(L) = \frac{\partial}{\partial t} \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \left( \langle n_I^*, t | \hat{H}_{0I}(t) + \hat{V}_I(t) | n_I^*, t \rangle - \langle n^* | \hat{H}_0 + \hat{V} | n^* \rangle \right) \quad (6.18)$$

where  $\mathcal{Z}^* = \sum_n e^{-\beta E_n^*}$  is the distribution function for static Hamiltonian  $\hat{H}_0 + \hat{V}$ ;  $|n_I^*, t\rangle = e^{-\frac{i}{\hbar} \int_{-\infty}^t \hat{H}'(t') dt'} |n^*\rangle$  is the interaction picture wavefunction, where  $\hat{H}'(t) = \sum_s \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)}$ ;  $\hat{H}'_I(t)$ ,  $\hat{V}_I(t)$  and  $\hat{H}_{0I}$  are interaction picture operators: for arbitrary operator  $\hat{A}(t)$  the interaction picture version is

$$\hat{A}_I(t) = e^{i(\hat{H}_0 + \hat{V})t/\hbar} \hat{A}(t) e^{-i(\hat{H}_0 + \hat{V})t/\hbar} \quad (6.19)$$

By expanding up to the second order in phonon external strain field  $e_{ij}(\vec{x}, t)$  there are four terms in total energy absorption rate Eq.(6.18). Three of them come from perturbation  $\hat{H}'(t)$ , the last one comes from non-elastic stress-stress interaction  $\hat{V}$ . We first consider the energy absorption rate due to perturbation  $\hat{H}'(t)$ . It contains three terms, one is quadratic in operator  $\sum_s \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)}$ , giving the energy absorption rate  $\dot{E}_{l,t}^{(1)}(L) = N_0^3 \dot{E}_{l,t}^{\text{single}}(L)$ . The second term is quadratic in the expectation value of

$$\sum_{ss'} \sum_{ijkl} \Delta \Lambda_{ijkl}^{(ss')} (t) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}:$$

$$\begin{aligned} \dot{E}_l^{(2)}(L) &= (1 - e^{-\beta\hbar\omega}) [(55 + 176\alpha + 688\alpha^2) + 44(1 + 4\alpha + 4\alpha^2)x(T, \omega)] \\ &\quad \frac{A^2 k^2 N_0^3 \ln N_0}{40\pi^3 (\rho c_t^2)^2} \omega \int \text{Im} \tilde{\chi}_t^{\text{res}}(T, \Omega) \text{Im} \tilde{\chi}_t^{\text{res}}(T, \omega - \Omega) d\Omega \\ \dot{E}_t^{(2)}(L) &= (1 - e^{-\beta\hbar\omega}) [(35 + 112\alpha + 656\alpha^2) + 28(1 + 4\alpha + 4\alpha^2)x(T, \omega)] \\ &\quad \frac{A^2 k^2 N_0^3 \ln N_0}{40\pi^3 (\rho c_t^2)^2} \omega \int \text{Im} \tilde{\chi}_t^{\text{res}}(T, \Omega) \text{Im} \tilde{\chi}_t^{\text{res}}(T, \omega - \Omega) d\Omega \end{aligned} \quad (6.20)$$

For details of calculations, please see Appendix (C). Please note, that the above two results are of the “quadratic order” of the imaginary part of resonance susceptibilities. So they are positive quantities.  $\alpha = 1 - \frac{c_t^2}{c_l^2}$  and  $x(T, \omega) = \frac{\text{Im} \tilde{\chi}_l^{\text{res}}(T, \omega)}{\text{Im} \tilde{\chi}_t^{\text{res}}(T, \omega)} - 2$ . Again we assume  $\text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega) \approx \text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T)$  is weakly dependent on frequency within a certain extent  $\omega < \omega_c$  for  $T < 1\text{K}$ . For details of discussions regarding  $\omega_c$ , please refer to section 4, Eq.(6.25). Eq.(6.20) are given by the convolution between  $\text{Im} \tilde{\chi}_t^{\text{res}}(T, \Omega)$  and  $\text{Im} \tilde{\chi}_t^{\text{res}}(T, \omega - \Omega)$ . By substituting qualitative first order expansion  $\Delta \hat{T}_{ij} \sim e \hat{T}_{ij} + \mathcal{O}(e^2)$  the energy absorption rate due to external phonon is

$$\dot{E}_{l,t}^{(3)}(L) \sim K_{l,t} (1 - e^{-\beta\hbar\omega}) \frac{A^2 k^2 N_0^3 \ln N_0}{\pi^3 (\rho c_t^2)^2} \omega \int \text{Im} \tilde{\chi}_t^{\text{res}}(T, \Omega) \text{Im} \tilde{\chi}_t^{\text{res}}(T, \omega - \Omega) d\Omega \quad (6.21)$$

For details of calculations, please see Appendix (C). The above result is also positive.  $K_{l,t}$  are constants for longitudinal and transverse cases, of order  $\sim 1$ . By comparing Eq.(6.20) and (6.21), the energy absorption from  $\Delta \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}$  and  $\Lambda_{ijkl}^{(ss')} \Delta \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}$  have the same scale dependence. In the next section we will demonstrate both of them are renormalization irrelevant at experimental length scale. Finally we consider the fourth contribution of energy absorption from non-elastic stress-stress interaction  $\hat{V}$ : by expanding it to the second order of  $e_{ij}(\vec{x}, t)$ , the energy absorption rate contribution from  $\hat{V}$  is given as follows

$$\dot{V}_{l,t}(L) = (1 - e^{-\beta\hbar\omega}) \frac{4N_0^3 L^3 A^2 k^2 \ln N_0}{\rho c_{t,l}^2} \omega \text{Im} \tilde{\chi}_{t,l}^{\text{res}}(T, \omega) \text{Re} \tilde{\chi}_{t,l}^{\text{res}}(T, \omega) \quad (6.22)$$

where we define “real part reduced non-elastic resonance susceptibility”,  $\text{Re} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega) = \frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\Omega \text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \Omega) d\Omega}{\Omega^2 - \omega^2}$ . For details of calculations, please see Appendix (C). Also please refer to Appendix (C) for the details of definitions of  $\text{Re} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega)$ . Since both of real and imaginary parts of resonance susceptibilities are negative quantities, the above many-body interaction’s contribution to energy absorption is positive. The total energy absorption of super block is given by the summation of the above four terms  $\dot{E}_{l,t}^{\text{super}}(L) =$

$\dot{E}_{l,t}^{(1)}(L) + \dot{E}_{l,t}^{(2)}(L) + \dot{E}_{l,t}^{(3)}(L) + \dot{V}_{l,t}(L)$ . Because “super block” at length scale  $L$  is the “single block” at length scale  $N_0L$ , we have the important relation  $\dot{E}_{l,t}^{\text{single}}(N_0L) = \dot{E}_{l,t}^{\text{super}}(L)$ . Super block energy absorption ratio due to longitudinal and transverse input phonon is therefore  $\dot{E}_l^{\text{single}}(N_0L)/\dot{E}_t^{\text{single}}(N_0L)$ . The Meissner-Berret ratio at length scale  $N_0L$  is  $\gamma_l^2/\gamma_t^2 = \dot{E}_l^{\text{single}}(N_0L)/\dot{E}_t^{\text{single}}(N_0L)$  which is different from that at length scale  $L$ ,  $\gamma_l^2/\gamma_t^2 = \dot{E}_l^{\text{single}}(L)/\dot{E}_t^{\text{single}}(L)$ . This implies that Meissner-Berret ratio is not a constant with the increase of length scale because of non-elastic stress-stress interaction. To study the universality of Meissner-Berret ratio we need to obtain energy absorption rate  $\dot{E}_{l,t}$  at experimental length scale  $R$ .

## 6.4 Renormalization Procedure of Susceptibility

In this section we want to get the energy absorption rate at experimental length scale by repeating renormalization procedure of combining single blocks into a super block. From the argument by D. C. Vural and A. J. Leggett[25] we start the renormalization procedure at length scale  $L_1 \sim 50\text{\AA}$ . Since the final result only logarithmically depends on this choice, it will not be sensitive. In the  $n$ -th step renormalization, we combine  $N_0^3$  single blocks with the dimension  $L_n \times L_n \times L_n$  to form a  $n$ -th step super block with the dimension  $N_0L_n \times N_0L_n \times N_0L_n$ . In the next step single block dimension is  $L_{n+1} = N_0L_n$ . By plugging in a weak phonon, the  $n$ -th step single and super block energy absorption rates are  $\dot{E}_{l,t}^{\text{single}}(L_n)$  and  $\dot{E}_{l,t}^{\text{super}}(L_n)$ . From the relation  $\dot{E}_{l,t}^{\text{super}}(L_n) = \dot{E}_{l,t}^{\text{single}}(L_{n+1})$  we get the following recursion of energy absorption rate from step  $n$  to  $n+1$ :

$$N_0^3 \dot{E}_{l,t}^{\text{single}}(L_n) + \dot{E}_{l,t}^{(2)}(L_n) + \dot{E}_{l,t}^{(3)}(L_n) + \dot{V}_{l,t}(L_n) = \dot{E}_{l,t}^{\text{single}}(L_{n+1}) \quad (6.23)$$

It is convenient to define “energy absorption rate per volume”:  $\dot{\epsilon}_{l,t}^{\text{single}}(L_n) = L_n^{-3} \dot{E}_{l,t}^{\text{single}}(L_n)$ ,  $\dot{\epsilon}_{l,t}^{(2,3)}(L_n) = L_{n+1}^{-3} \dot{E}_{l,t}^{(2,3)}(L_n)$ ,  $\dot{v}_{l,t}(L_n) = L_{n+1}^{-3} \dot{V}_{l,t}(L_n)$  and  $\dot{\epsilon}_{l,t}^{\text{single}}(L_{n+1}) = L_{n+1}^{-3} \dot{E}_{l,t}^{\text{single}}(L_{n+1})$ . Repeat renormalization procedure  $\log_{N_0}(R/L_1)$  times from unit block length scale  $L_1 \sim 50\text{\AA}$  to experimental length scale  $R$ , the energy absorption rate per volume is

$$\dot{\epsilon}_{l,t}(R) = \left( \dot{\epsilon}_{l,t}(L_1) + \dot{\epsilon}_{l,t}^{(2)}(L_1) + \dot{\epsilon}_{l,t}^{(3)}(L_1) \right) + \dot{v}_{l,t} \log_{N_0} \left( \frac{R}{L_1} \right) \quad (6.24)$$

First of all we compare the volume dependence of  $\dot{\epsilon}_{l,t}^{(2)}(L)$ ,  $\dot{\epsilon}_{l,t}^{(3)}(L)$  and  $\dot{v}_{l,t}$ :

$$\begin{aligned} \frac{\dot{\epsilon}_{l,t}^{(2,3)}(L)}{\dot{v}_{l,t}} &= \frac{1}{\rho c_{l,t}^2 L^3} \frac{\int \text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \Omega) \text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega - \Omega) d\Omega}{\text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega) \int \frac{\Omega \text{Im} \tilde{\chi}_{l,t}^{\text{res}}(\Omega) d\Omega}{\Omega^2 - \omega^2}} \sim \frac{1}{\rho c_{l,t}^2 L^3} \frac{\omega_c}{\int^{\omega_c} \Omega d\Omega / (\Omega^2 - \omega^2)} \\ &\sim \frac{1}{\rho c_{l,t}^2 L^3} \frac{\omega_c}{\ln(\omega_c/\omega)} \end{aligned} \quad (6.25)$$

where  $L$  is length scale. In the above result we use the assumption that susceptibility  $\text{Im} \tilde{\chi}_{l,t}(T, \omega)$  is roughly a constant of frequency for  $\omega < \omega_c$  below temperature 1K.  $\dot{\epsilon}_{l,t}^{(2,3)}(L)$  and  $\dot{v}_{l,t}$  have the same unit (energy per volume per unit time), however the upper limit of integrals  $\omega_c$  in Eq.(6.25) does not increase with the increase  $L$ ,  $\dot{\epsilon}_{l,t}^{(2)}(L)$  and  $\dot{\epsilon}_{l,t}^{(3)}(L)$  are  $L^{-3}$  volume dependent while  $\dot{v}_{l,t}$  is scale invariant. With the increase of length scale eventually  $\dot{v}_{l,t}$  will be greater than  $\dot{\epsilon}_{l,t}^{(2,3)}(L)$  beyond critical length  $L_c$ . We use ultrasonic frequency  $\omega \sim 10^6$ rad/s, amorphous solid mass density  $\rho \sim 10^3$ kg/m<sup>3</sup> and speed of sound  $c \sim 10^3$ m/s to estimate the critical length scale when  $\dot{\epsilon}_{l,t}^{(2,3)}(L)$  and  $\dot{v}_{l,t}$  become comparable. The upper limit of  $\omega_c$  is of order  $10^{15}$ rad/s corresponding to temperature  $10^4$ K, so the largest possible  $L_c$  is of order  $\sim 10\text{\AA}$ , even smaller than starting length of renormalization procedure  $L_1 \sim 50\text{\AA}$ . Therefore throughout the entire process of renormalization,  $\dot{\epsilon}_{l,t}^{(2,3)}(L)$  is always negligible compared to  $\dot{v}_{l,t}$ . We conclude  $\dot{\epsilon}_{l,t}^{(2,3)}(L_1)$  is renormalization irrelevant in Eq.(6.24).

Next let us compare the renormalization relevance between  $\dot{\epsilon}_{l,t}(L_1)$  and  $\dot{v}_{l,t} \log_{N_0}(R/L_1)$ . The input ultrasonic phonon frequency usually takes the order  $\sim 10^6$ Hz, corresponding to wavelength  $R \sim 10^{-3}$ m. Therefore the experimental length scale  $R$  is the wavelength of external phonon, because it is smaller than the actual size of amorphous samples. With this choice  $\ln N_0 \log_{N_0}(R/L_1) = \ln(R/L_1) \sim 20 \gg 1$ , so we assume the energy absorption rate per volume of unit block  $\dot{\epsilon}_{l,t}(L_1)$  is much smaller than that from stress-stress interactions  $\dot{v}_{l,t} \log_{N_0}(R/L_0)$ . At experimental length scale the energy absorption rate is dominated by  $\dot{v}_{l,t}$ , independent of material microscopic nature. The ratio between longitudinal and transverse energy absorption rate per volume at experimental length scale is given as follows:

$$\frac{\dot{\epsilon}_l(R)}{\dot{\epsilon}_t(R)} = \frac{\dot{v}_l}{\dot{v}_t} \quad (6.26)$$

Note the r.h.s. of Eq.(6.26) is functional of  $\text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega)$ ; for the l.h.s., at experimental length scale the entire amorphous sample can be treated as a huge block, with the energy absorption rate per volume  $\dot{\epsilon}_{l,t}(R) = 2A^2 k^2 \omega (1 - e^{-\beta \hbar \omega}) \text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega)$ . So the l.h.s. of Eq.(6.26) equals to  $\frac{\text{Im} \tilde{\chi}_l^{\text{res}}(T, \omega)}{\text{Im} \tilde{\chi}_t^{\text{res}}(T, \omega)}$ . Eq.(6.26) together with Eq.(6.22) is a self-consistent equation for  $\text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega)$ . The only parameter enters it is speed of sound ratio  $c_l/c_t$  and it is a non-adjustable quantity. The self-consistent equation for Meissner-Berret

ratio  $\sqrt{\text{Im } \tilde{\chi}_l^{\text{res}}(T, \omega) / \text{Im } \tilde{\chi}_t^{\text{res}}(T, \omega)}$  turns out to be

$$\frac{\text{Im } \tilde{\chi}_l^{\text{res}}(T, \omega)}{\text{Im } \tilde{\chi}_t^{\text{res}}(T, \omega)} = \frac{c_t^2 \text{Im } \tilde{\chi}_l^{\text{res}}(T, \omega) \text{Re } \tilde{\chi}_l^{\text{res}}(T, \omega)}{c_l^2 \text{Im } \tilde{\chi}_t^{\text{res}}(T, \omega) \text{Re } \tilde{\chi}_t^{\text{res}}(T, \omega)} \Rightarrow \frac{\text{Re } \tilde{\chi}_l^{\text{res}}(T, \omega)}{\text{Re } \tilde{\chi}_t^{\text{res}}(T, \omega)} = \frac{c_l^2}{c_t^2} \quad (6.27)$$

Eq.(6.27) only tells us that the ratio between real part of non-elastic resonance susceptibility is sound velocity ratio squared. But please note that the ratio between  $\text{Re } \tilde{\chi}_l^{\text{res}}(T, \omega)$  and  $\text{Re } \tilde{\chi}_t^{\text{res}}(T, \omega)$  turns out to be always  $c_l^2/c_t^2$ , regardless of their frequency. We can obtain the imaginary part susceptibility ratio via Kramers-Kronig relation from this frequency-independent property:

$$\text{Im } \tilde{\chi}_{l,t}^{\text{res}}(T, \omega) = -\frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\omega \text{Re } \tilde{\chi}_{l,t}^{\text{res}}(T, \Omega)}{\Omega^2 - \omega^2} d\Omega \Rightarrow \frac{\text{Im } \tilde{\chi}_l^{\text{res}}(T, \omega)}{\text{Im } \tilde{\chi}_t^{\text{res}}(T, \omega)} = \frac{c_l^2}{c_t^2} \quad (6.28)$$

Finally we get the ratio between longitudinal and transverse reduced version of imaginary non-elastic susceptibilities:  $\sqrt{\text{Im } \tilde{\chi}_l^{\text{res}}(T) / \text{Im } \tilde{\chi}_t^{\text{res}}(T)} = c_l/c_t$  (and so as  $\sqrt{\text{Im } \chi_l^{\text{res}}(T) / \text{Im } \chi_t^{\text{res}}(T)} = c_l/c_t$ ). On the other hand by comparing TTLS energy absorption  $\dot{E}_{l,t} \propto \gamma_{l,t}^2$  we have  $\sqrt{\text{Im } \tilde{\chi}_l^{\text{res}}(T) / \text{Im } \tilde{\chi}_t^{\text{res}}(T)} = \gamma_l/\gamma_t$ , so theoretical Meissner-Berret ratio  $\gamma_l/\gamma_t = c_l/c_t$ .

This result is in fairly good agreement with 13 materials we list below. Experimental coupling constants  $\gamma_{l,t}$ , Meissner-Berret ratio  $(\gamma_l/\gamma_t)^{\text{exp}}$  and speed of sound  $c_{l,t}$  are from the data by Meissner and Berret[45];  $(\gamma_l/\gamma_t)^{\text{theo}}$  is our self-consistent result:

Material	$\gamma_l(\text{eV})$	$\gamma_t(\text{eV})$	$(\gamma_l/\gamma_t)^{\text{exp}}$	$c_l(\text{km/s})$	$c_t(\text{km/s})$	$(\gamma_l/\gamma_t)^{\text{theo}} = c_l/c_t$	$\frac{\text{theo-exp}}{\text{exp}}$
a-SiO <sub>2</sub>	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 <sub>2</sub> S <sub>3</sub>	0.26	0.17	1.53	2.70	1.46	1.85	+20.9%
LaSF-7	1.46	0.92	1.59	5.64	3.60	1.57	-1.26%
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	0.39	0.27	1.44	3.15	1.57	2.01	+39.6%
PS	0.20	0.13	1.54	2.80	1.50	1.87	+21.4%

Among 13 materials, the theoretical results of  $\text{As}_2\text{S}_3$ , PMMA and PS deviate more than 20% compared to their experimental measurements. We give below a further discussion on these three materials. For now let's investigate the statistical significance between theoretical and experimental Meissner-Berret ratio. We use least square method. For 13 materials including large deviations of  $\text{As}_2\text{S}_3$ , PMMA and PS, the fitted linear relation is  $\left(\frac{\gamma_l}{\gamma_t}\right)^{\text{theo}} = 1.102 \left(\frac{\gamma_l}{\gamma_t}\right)^{\text{exp}}$  with the correlation coefficient  $r = 0.261$ , which means linear fitting is not good for them; for 10 materials excluding  $\text{As}_2\text{S}_3$ , PMMA and PS, the fitted linear relation is  $\left(\frac{\gamma_l}{\gamma_t}\right)^{\text{theo}} = 1.061 \left(\frac{\gamma_l}{\gamma_t}\right)^{\text{exp}}$  with the correlation coefficient  $r = 0.745$ , which means except for large deviations  $\text{As}_2\text{S}_3$ , PMMA and PS,  $c_l/c_t$  is a moderate fitting for other 10 materials. We plot these data as follows, where  $x$  and  $y$ -axis represent experimental and theoretical Meissner-Berret ratio:

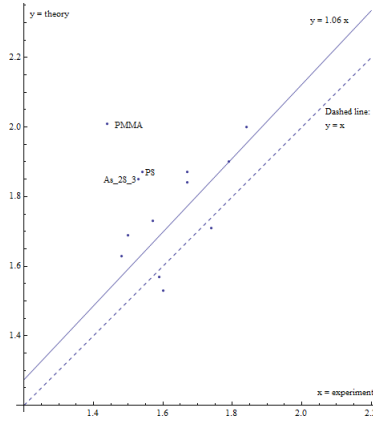


Figure 6.1: Least square fitting for experimental-theoretical Meissner-Berret ratio. The linear fitting is  $y = 1.06x$ ; correlation coefficient  $r = 0.261$  for 13 materials' data;  $r = 0.745$  for 10 materials' data excluding PMMA, PS and  $\text{As}_2\text{S}_3$ . The dashed line is our anticipation on theory  $(\gamma_l/\gamma_t)^{\text{exp}} = c_l/c_t$ .

Instead of resonance energy absorption measurements, the original experiment of Meissner-Berret ratio[45] was to measure relative speed of sound shift to temperature,  $\Delta c_{l,t}/c_{l,t} = C_{l,t} \ln(T/T_0)$ , where the experimental measured constant  $C_{l,t}$  is derived by TTLS parameters  $C_{l,t} = \bar{P}\gamma_{l,t}^2/\rho c_{l,t}^2$ . The definition of  $\bar{P}$  is[10]: in TTLS model the diagonal matrix element  $\Delta$  and tunneling parameter  $\lambda = \ln(\hbar\Omega/\Delta_0)$  are assumed to be independent of each other and to have a constant distribution  $P(\Delta, \lambda)d\Delta d\lambda = \bar{P}d\Delta d\lambda$ . By measuring  $C_{l,t}$ ,  $c_{l,t}$  and  $\bar{P}$  one can experimentally calculate coupling constants  $\gamma_{l,t}$ . However, it may not always be true that  $\Delta, \lambda$  exactly obeys constant distribution. We search experimental data for low-temperature specific heat:  $\text{As}_2\text{S}_3$  measured by R. B. Stephens[48]; PMMA and PS measured by R. B. Stephens, G. S. Cieloszyk and G. L. Salinger[49]; PMMA measured by R. C. Zeller and R. O. Pohl[1]. At temperatures  $T < 1\text{K}$  their heat capacity temperature dependences largely deviate from  $C_v(T) = AT + BT^3$ , where  $A$  and  $B$  are experimentally determined parameters. Their huge deviations from “linear temperature dependence” implies that TTLS assumptions may not be a suitable description below 1K, especially when measuring quantities

sensitive to distribution constant  $\bar{P}$  like  $\gamma_{l,t}$ . We think this might be one of the reasons why theoretical Meissner-Berret ratio  $\gamma_l/\gamma_t$  for  $\text{As}_2\text{S}_3$ , PMMA and PS deviate more than 20% from experimental data.

We also suggest the possibility that  $\text{As}_2\text{S}_3$ , PMMA and PS may not possess universal low-temperature glass properties observed in typical glass materials (e.g.  $\text{a-SiO}_2$ ,  $(\text{KCl})_x(\text{KCN})_{1-x}$  etc.): for example, quadratic temperature dependence of thermal conductivity  $\kappa \sim T^2$  below  $T < 1\text{K}$ [48]; universal thermal conductivity plateau between 4K and 20K[48]; universal sound velocity shift discussed in chapter 4[33], and so on. We hope more experiments of low-temperature thermal and acoustic properties on these three materials could be carried out to test our predictions.

## 6.5 The Modification of Meissner-Berret Ratio from Electric Dipole-Dipole Interactions

Electric dipole moments interact with each other via  $r^{-3}$  long range interaction similar with non-elastic stress-stress interactions. In this section we take electric dipole moments as operators which interact like non-elastic stress stress interactions. The input mechanical waves (not electromagnetic waves) can change the relative positions  $\vec{x}_s - \vec{x}_{s'}$  of dipole moments at  $\vec{x}_s$  and  $\vec{x}_{s'}$ ; on the other hand, electric dipole moment is proportional to the separation of positive-negative charges:  $\vec{p} = q\vec{l}$ . Thus external phonons also modify dipole moments by changing charge separation  $\vec{l} \rightarrow \vec{l} + \Delta\vec{l}$ . Finally, external phonons will change electric dipole interactions, resulting in the change of amorphous material energy absorption.

However, as we will see at the end of this section, the influence of electric dipole-dipole interaction on phonon energy absorption is renormalization irrelevant, because of the following reason. From section 3, we know the non-elastic stress-stress interaction  $\hat{V}$  has four contributions to the resonant phonon energy absorption rate  $\dot{E}_{l,t}$ : Eqs.(6.20), Eq.(6.21) and Eq.(6.22). Eqs.(6.20) and Eq.(6.21) are renormalization irrelevant, while Eq.(6.22) is the only renormalization relevant term for phonon energy absorption. Let us give a short review on Eqs.(6.20, 6.21, 6.22): Eq.(6.20) is generated by the change of non-elastic stress-stress interaction coefficient  $\Delta\Lambda_{ijkl}^{(ss')}(\mathbf{e}) = \Lambda_{ijkl}^{(ss')}(\mathbf{e}) - \Lambda_{ijkl}^{(ss')}$  due to external strain field; Eq.(6.21) is generated by the change of non-elastic stress tensor operator  $\Delta\hat{T}_{ij} = \hat{T}_{ij}(\mathbf{e}) - \hat{T}_{ij}$ . These two terms generate phonon energy absorptions which are renormalization irrelevant; Eq.(6.22) is generated by the single block wave function change  $\delta|n\rangle$  due to the coupling between stress tensor and strain field  $e_{ij}\hat{T}_{ij}$ . This term is renormalization relevant. When we consider the resonant phonon energy absorption contribution from electric dipole-dipole interaction, we get the following contributions: (1) electric dipole-dipole interaction coefficient  $\mu_{ij}^{(ss')}$  could be modified by external phonon field, thus we have the change of interaction coefficient  $\Delta\mu_{ij}^{(ss')} = \mu_{ij}^{(ss')}(\mathbf{e}) -$



$\mu_{ij}^{(ss')}$ . The contribution to phonon energy absorption from this term is renormalization irrelevant, which is similar with Eq.(6.20); (2) electric dipole moments  $\hat{p}_i$  could be changed by external phonon strain field, resulting in the change  $\Delta\hat{p}_i = \hat{p}_i(\mathbf{e}) - \hat{p}_i$ . The phonon energy absorption contribution from this term is also renormalization irrelevant, similar to Eq.(6.21); (3) while elastic strain field can couple to non-elastic stress tensor with  $e_{ij}\hat{T}_{ij}$  and electric field can couple to electric dipole moment with  $-E_i\hat{p}_i$ , there is not a term, that elastic strain field can couple to electric dipole moment. The phonon resonant energy absorption from the coupling  $e_{ij}\hat{T}_{ij}$  is the renormalization relevant term, which does not exist in electric dipole moments with the coupling to external phonon strain field.

Let's first qualitatively compare the order of magnitude for the influence of phonon energy absorption between electric dipole interaction and non-elastic stress-stress interaction. We use  $M$  to denote the value of off-diagonal matrix element for two-level-system and use  $n_0$  to denote the density of states for TTLS system. We also use  $\mu$  to denote the electric dipole moment and use  $n_e$  to denote the density of states for electric dipole two-level-system. In the theory of tunneling-two-level-system model, the quantity  $\frac{n_0 M^2}{\rho c^2}$  represents the average of the imaginary part of non-elastic resonance susceptibility of two-level-system divided by  $\rho c^2$ , while  $\frac{n_e \mu^2}{\epsilon}$  represents the average of the imaginary part of dielectric resonance susceptibility divided by  $\epsilon$ . On the other hand, the quantities  $\frac{n_0 M^2}{\rho c^2}$  and  $\frac{n_e \mu^2}{\epsilon}$  correspond to  $\frac{\text{Im} \chi_t}{\rho c_t^2}$  and  $\frac{\text{Im} \chi}{\epsilon}$  in our generic coupled block model. Since the ‘‘resonant phonon energy absorption rate’’  $\dot{E}_{l,t}$  is proportional to  $\left(\frac{\text{Im} \chi_{l,t}}{\rho c_{l,t}^2}\right)^2$  via non-elastic stress-stress interaction, and is proportional to  $\left(\frac{\text{Im} \chi}{\epsilon}\right)^2$  via electric dipole-dipole interaction (we will prove this below), the ratio between  $\frac{n_0 M^2}{\rho c^2}$  and  $\frac{n_e \mu^2}{\epsilon}$  gives us the order of magnitude comparison on phonon energy absorption rate via non-elastic stress-stress interaction and electric dipole-dipole interaction. With the measurement from S. Hunklinger and M. V. Schickfus[46] we discuss the ratio between  $\frac{n_0 M^2}{\rho c^2}$  and  $\frac{n_e \mu^2}{\epsilon}$  for two dielectric materials, BK7 and SiO<sub>2</sub> below.

For BK7, TTLS parameters are of order  $n_0 M^2 \sim 10^8 \text{erg/cm}^3$ ; dielectric constant  $\epsilon = 3.7$ ;  $n_e \mu^2 = 6 \times 10^{-3}$ ; mass density  $\rho = 2.51 \text{g/cm}^3$ ; speed of sound  $c = 6.5 \times 10^5 \text{cm/s}$ . From these data we find the following ratio between electric dipole-dipole interaction and non-elastic stress-stress interaction is  $(n_e \mu^2 / \epsilon : n_0 M^2 / \rho c^2) \sim (1.62 \times 10^{-3} : 0.94 \times 10^{-4})$ , which means the influence on phonon energy absorption due to electric dipole interaction is one order of magnitude greater than that of non-elastic stress-stress interaction for BK7.

For SiO<sub>2</sub>, TTLS parameters  $n_0 M^2 = 2.04 \times 10^8 \text{erg/cm}^3$ ;  $\epsilon = 3.81$ ; electric dipole moment parameters  $n_e \mu^2 = 1.46 \times 10^{-4}$ ;  $\rho = 2.2 \text{g/cm}^3$ ;  $c = 5.8 \times 10^5 \text{cm/s}$ ; the strength of electric dipole interaction versus non-elastic stress-stress interaction is  $(n_e \mu^2 / \epsilon : n_0 M^2 / \rho c^2) \sim (3.83 \times 10^{-5} : 2.76 \times 10^{-4})$ , which means for SiO<sub>2</sub>, the influence of phonon energy absorption due to electric dipole interaction is one order of magnitude smaller than that of non-elastic stress-stress interaction.

The above qualitative arguments suggest electric dipole interaction in dielectric materials is roughly of the same order of non-elastic stress-stress interactions. However, after a detailed calculation we demonstrate the energy absorption contribution of electric dipole interaction is renormalization irrelevant. 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, when  $\vec{x}_s$  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 for  $s$ -th block. By combining  $N_0 \times N_0 \times N_0$  identical  $L \times L \times L$  unit blocks to form a  $N_0 L \times N_0 L \times N_0 L$  super block, the electric dipole interaction

$$\hat{V}_{\text{dipole}} = \sum_{s \neq s'}^{N_0^3} \sum_{i,j=1}^3 \mu_{ij}^{(ss')} \hat{p}_i^{(s)} \hat{p}_j^{(s')} \quad (6.29)$$

in the above equation we define the coefficient  $\mu_{ij}^{(ss')}$

$$\mu_{ij}^{(ss')} = \frac{(\delta_{ij} - 3n_i n_j)}{8\pi\epsilon |\vec{x}_s - \vec{x}_{s'}|^3} \quad (6.30)$$

in Eq.(6.29, 6.30)  $i, j$  runs over 1, 2, 3 cartesian coordinates and  $\vec{n}$  is the unit vector of  $\vec{x}_s - \vec{x}_{s'}$ . The input phonon field  $\vec{u}(\vec{x}, t)$  can modify (1) dipole interaction coefficient  $\mu_{ij}^{(ss')}$  by changing relative positions of blocks  $\vec{x}_s - \vec{x}_{s'}$ . We denote it  $\Delta\mu_{ij}^{(ss')}$ :

$$\Delta\mu_{ij}^{(ss')} = \frac{3\Delta x_{ss'}}{8\pi\epsilon x_{ss'}^4} [(5n_i n_j - \delta_{ij}) \cos \theta_{ss'} - (n_j m_i + n_i m_j)] \quad (6.31)$$

where  $x_{ss'} = |\vec{x}_s - \vec{x}_{s'}|$ ,  $\Delta\vec{x}_s = \vec{u}(\vec{x}_s, t)$ ,  $\Delta x_{ss'} = |\Delta\vec{x}_s - \Delta\vec{x}_{s'}|$ ,  $\cos \theta_{ss'} = (\Delta\vec{x}_{ss'} \cdot \vec{x}_{ss'}) / \Delta x_{ss'} x_{ss'}$  and  $\vec{m} = \Delta\vec{x}_{ss'} / \Delta x_{ss'}$ . (2) Phonon field can also change dipole operators  $\hat{p}^{(s)}$ , because positive negative charges in electric dipole are driven from original positions  $\vec{x}_s \pm \frac{1}{2}\vec{l}_s$  to new positions  $\vec{x}_s \pm \frac{1}{2}\vec{l}_s + \vec{u}(\vec{x}_s \pm \frac{1}{2}\vec{l}_s, t)$ , leading to the change of dipole operators  $\Delta\hat{p}^{(s)}$

$$\Delta\hat{p}_i(\vec{x}, t) = \sum_k \frac{\partial u_i(\vec{x}, t)}{\partial x_k} \hat{p}_k(\vec{x}) \quad (6.32)$$

Therefore with the presence of external phonon field the total electric dipole interaction is given by

$$\hat{V}_{\text{dipole}}(\mathbf{e}) = \sum_{s \neq s'}^{N_0^3} \sum_{i,j=1}^3 \left( \mu_{ij}^{(ss')} \hat{p}_i^{(s)} \hat{p}_j^{(s')} + \Delta\mu_{ij}^{(ss')}(t) \hat{p}_i^{(s)} \hat{p}_j^{(s')} + 2\mu_{ij}^{(ss')} \Delta\hat{p}_i^{(s)}(t) \hat{p}_j^{(s')} \right) \quad (6.33)$$

Let's define electric dipole-dipole susceptibility  $\chi_{ij}(T, \omega)$  for future use:

$$\begin{aligned}
\text{Im}\chi_{ij}(T, \omega) &= (1 - e^{-\beta\hbar\omega}) \text{Im}\tilde{\chi}_{ij}(T, \omega) \\
\text{Im}\tilde{\chi}_{ij}(T, \omega) &= \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \text{Im}\tilde{\chi}_{ij}^{(m)}(\omega) \\
\text{Im}\tilde{\chi}_{ij}^{(m)}(\omega) &= \frac{\pi}{L^3} \sum_n \langle m|\hat{p}_i^{(s)}|n\rangle \langle n|\hat{p}_j^{(s)}|m\rangle [-\delta(E_n - E_m - \omega)]
\end{aligned} \tag{6.34}$$

Since the dipole-dipole susceptibility must be invariant under SO(3) group transformations, it takes the generic form  $\text{Im}\tilde{\chi}_{ij}(T, \omega) = \text{Im}\tilde{\chi}(T, \omega)\delta_{ij}$ .

To consider energy absorption we follow two steps: (1) turn off stress-stress interaction  $\hat{V}$  and dipole interaction  $\hat{V}_{\text{dipole}}$ . These  $N_0^3$  non-interacting blocks' Hamiltonian is  $\hat{H}_0 = \sum_s \hat{H}_0^{(s)}$ . We denote  $|n\rangle = \prod_s |n^{(s)}\rangle$  and  $E_n = \sum_s E_n^{(s)}$  to be the eigenstates and eigenvalues for  $\hat{H}_0$ ; (2) turn on time-dependent perturbation

$$\begin{aligned}
\hat{H}'(t) &= \sum_s \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)} + \sum_{ss'} \sum_{ijkl} \left( \Delta\Lambda_{ijkl}^{(ss')} (t) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} + 2\Lambda_{ijkl}^{(ss')} \Delta\hat{T}_{ij}^{(s)}(t) \hat{T}_{kl}^{(s')} \right) \\
&+ \sum_{ss'} \sum_{ij} \left( \Delta\mu_{ij}^{(ss')} (t) \hat{p}_i^{(s)} \hat{p}_j^{(s')} + 2\mu_{ij}^{(ss')} \Delta\hat{p}_i^{(s)}(t) \hat{p}_j^{(s')} \right)
\end{aligned} \tag{6.35}$$

and static interaction  $\hat{V} + \hat{V}_{\text{dipole}}$  to consider energy absorption of super block Hamiltonian  $\hat{H}_0 + \hat{V} + \hat{V}_{\text{dipole}}$ :

$$\dot{E}_{l,t}^{\text{super}}(L) = \partial_t \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \left( \langle n_I, t | \hat{H}_0 + \hat{V}_I(t) + \hat{H}'_I(t) | n_I, t \rangle - \langle n | \hat{H}_0 + \hat{V} | n \rangle \right) \tag{6.36}$$

with  $|n_I, t\rangle = e^{-\frac{i}{\hbar} \int_{-\infty}^t \hat{H}'_I(t') dt'} |n\rangle$ , and  $\hat{H}'_I(t)$  and  $\hat{V}_I(t)$  are interaction picture wavefunction and operators we discussed previously. Besides the energy absorption terms we have obtained in Eq.(6.20), Eq.(6.21) and Eq.(6.22), there is one extra term from electric dipole interactions. Expand the extra contribution to energy absorption in orders of  $e_{ij}(\vec{x}, t)$  the first order vanishes; the second order expansion is

$$\begin{aligned}
\dot{E}_l^{\text{dipole}} &= \frac{94A^2 k^2 N_0^3 \ln N_0}{960\pi^2 \epsilon^2} (1 - e^{-\beta\hbar\omega}) \omega \int \text{Im}\tilde{\chi}(T, \Omega) \text{Im}\tilde{\chi}(T, \omega - \Omega) d\Omega \\
\dot{E}_t^{\text{dipole}} &= \frac{53A^2 k^2 N_0^3 \ln N_0}{960\pi^2 \epsilon^2} (1 - e^{-\beta\hbar\omega}) \omega \int \text{Im}\tilde{\chi}(T, \Omega) \text{Im}\tilde{\chi}(T, \omega - \Omega) d\Omega
\end{aligned} \tag{6.37}$$

Next we need to plug the above contribution, Eq.(6.37) into the renormalization procedure of phonon resonant energy absorption between different length scales, Eq.(6.23). We repeat the RG steps, and obtain experimental length scale self-consistent equation for Meissner-Berret ratio  $\gamma_l/\gamma_t = \sqrt{\text{Im}\tilde{\chi}_l(T)/\text{Im}\tilde{\chi}_t(T)}$ . However, let's stop for the moment and discuss the scale dependence of Eq.(6.37). It is convenient to define

“energy absorption per volume”  $\dot{\epsilon}_{l,t}^{\text{dipole}} = (N_0 L)^{-3} \dot{E}_{l,t}^{\text{dipole}}$ . From the qualitative order of magnitude analysis,  $\dot{\epsilon}_{l,t}^{\text{dipole}}$  has the same order of magnitude as  $\dot{\epsilon}_{l,t}^{(2,3)}$  (see Eq.(6.20, 6.21) divided by volume). It also has the same volume dependence as  $\dot{\epsilon}_{l,t}^{(2,3)}$ . To illustrate this property, let us compared the quantities  $\dot{\epsilon}_{l,t}^{\text{dipole}}$  and  $\dot{v}_{l,t}$  (see Eq.(6.22) divided by volume) as follows

$$\frac{\dot{\epsilon}_{l,t}^{\text{dipole}}}{\dot{v}_{l,t}} \approx \frac{\frac{(\text{Im } \tilde{\chi})^2}{\epsilon^2} \omega_c}{L^3 \frac{(\text{Im } \tilde{\chi}_t)^2}{\rho c_{l,t}^2} \ln(\omega_c/\omega)} \quad (6.38)$$

where in the above result we assume dipole-dipole susceptibility  $\text{Im } \tilde{\chi}(T, \omega) \approx \text{Im } \tilde{\chi}(T)$  is roughly a constant of frequency within a certain range  $\omega < \omega_c$ . Eq.(6.38) indicates that the term  $\dot{\epsilon}_{l,t}^{\text{dipole}}$  is inversely proportional to  $L^3$  when compared to  $\dot{v}_{l,t}$ . Since  $\dot{\epsilon}_{l,t}^{\text{dipole}}$  decreases cubically with the increase of length scale, we assume beyond the critical length scale  $L_c = \left( \frac{(\text{Im } \tilde{\chi})^2 \rho c_{l,t}^2 \omega_c}{\epsilon^2 (\text{Im } \tilde{\chi}_t)^2 \ln(\omega_c/\omega)} \right)^{1/3}$ ,  $\dot{v}_{l,t}$  becomes much greater than  $\dot{\epsilon}_{l,t}^{\text{dipole}}$ . The upper limit of  $L_c$  can be obtained by letting  $\omega_c$  to take an extremely high value,  $\omega_c \sim 10^{15} \text{rad/s}$ . We obtain  $L_c \sim 10 \text{\AA}$ , even smaller than the starting length scale of real space renormalization procedure  $50 \text{\AA}$ . Throughout the entire renormalization procedure the influence of electric dipole-dipole interaction on phonon resonant energy absorption is always negligible.

# Chapter 7

## Conclusions

In this thesis we develop a generic coupled block model to explore three universal properties of low-temperature glass. They are: universal shift on glass sound velocity and dielectric constant, mechanical avalanche phenomena and universal Meissner-Berret ratio. The assumption we specify in this model is the correlation function (susceptibility) between non-elastic stress tensors and electric dipole moments are diagonal in spacial coordinates:  $\langle \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \rangle = \chi_{ijkl} \delta_{ss'}$ ,  $\langle \hat{p}_i^{(s)} \hat{p}_j^{(s')} \rangle = \chi_{ij} \delta_{ss'}$ . The exchange of virtual phonon and photon allows  $1/r^3$  long-range interactions. With the increase of system size, the increasing number of unit blocks compensate  $1/r^3$  decreasing behavior. The number of interactions increases quadratically with unit block numbers, while the number of single block Hamiltonian is proportional to it. Eventually at large length scale many body interaction dominate glass Hamiltonian. We use renormalization technique to iterate non-elastic and dielectric susceptibilities from small length scale to experimental length scale.

We want to set up a generic glass model to prove the universal slope ratio of temperature dependence on sound velocity shift, in relaxation and resonance regimes. We hope our renormalization technique would lead to the universal shift of sound velocity and dielectric constant, but in fact the renormalization equations in chapter 4 lead to the increasing behavior of relaxation and resonance susceptibilities rather than the expected decreasing behavior as the length scale increases. Moreover, the fixed point which gives the relation between relaxation and resonance susceptibilities at experimental length scale,  $\chi^{\text{rel}} = -2\chi^{\text{res}}(\omega = 0)$  can never be reached, due to the fact that both of relaxation and zero-frequency resonance susceptibilities are negative — they will always have the same sign throughout the entire renormalization process. It is at this point that our renormalization technique cannot explain the universal sound velocity and dielectric constant shift.

The second goal of this thesis is to use our generic coupled block model to understand the mechanical avalanche behavior of three-dimensional insulating glass. The reader should be aware that it is the first time to apply our model in glass mechanical avalanche problem. Therefore our purpose is not to solve the entire glass avalanche problem from microscopic point of view; instead we want to provide some first-step results for future people to continue studying this problem. We consider a block of amorphous material under the deformation of static, uniform strain. With the slowly increasing strain the bulk glass behaves elastically

until it reaches critical strain value. After that the stress ( $\mathbf{T}$ ) suddenly drops to a lower value. A more convenient quantity is the mechanical stress-stress susceptibility  $\chi_{ijkl} = \delta T_{ij} / \delta e_{kl}$ . At critical strain field when irreversible process happens, stress-stress susceptibility presents an abrupt positive-negative transition when strain field passes through critical value. Our main goal is to prove the existence of such positive-negative transition, and to obtain the exact value of critical strain value when avalanche happens. However, since the elastic susceptibility  $-(\rho c_l^2 - 2\rho c_t^2)\delta_{ij}\delta_{kl} - \rho c_t^2(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$  does not show such kind of positive-negative transition (which means it does not have a singularity), and the non-elastic susceptibility keeps negative throughout the entire renormalization procedure (which means it does not have a singularity as well), it is impossible to find a singularity in glass total mechanical susceptibility. It is at this point that our theory cannot explain the glass mechanical avalanche problem.

Among 13 materials measured by Meissner and Berret[45], 10 of them agree fairly good with theoretical results while other 3 are not. At first we thought this is because of huge electric dipole interactions. However qualitative measurements from Thomas, Ravindran and Varma[47] indicate that electric dipole interaction is too weak to affect Meissner-Berret ratio. We believe their huge deviations come from the reason that experimental data  $\gamma_l/\gamma_t$  were inferred from TTLS parameters. R. B. Stephens[48], G. S. Cieloszyk, G. L. Salinger[49], R. C. Zeller and R. O. Pohl[1]'s measurements on heat capacity indicate that constant parameter distribution for  $\text{As}_2\text{S}_3$ , PS and PMMA may not be a suitable description below 1K, so  $(\gamma_l/\gamma_t)^{\text{exp}}$  inferred from TTLS parameters for them may deviate from their original natures.

# Appendix A

## Derivation Details of Non-Elastic Stress-Stress Interaction Coefficient

It was Joffrin and Levelut[42] who firstly gave the detailed derivation of amorphous solid non-elastic stress-stress interaction coefficient  $\Lambda_{ijkl}^{(ss')}$ . We give a further correction to their results. To compare their result with ours, let us denote  $\left(\Lambda_{ijkl}^{(ss')}\right)_{\text{Joffrin}}$  for their stress-stress interaction coefficient :

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

where  $\alpha = 1 - c_t^2/c_l^2$ . We consider long wavelength limit. We will derive  $\Lambda_{ijkl}^{(ss')}$  starting from amorphous solid Hamiltonian written in the summation of phonon part, phonon-stress tensor coupling and non-elastic part of Hamiltonian:

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

where  $\mu$  is phonon polarization, i.e., longitudinal and transverse;  $\vec{q}$  is momentum and  $m$  the mass of elementary block,  $p_\mu(\vec{q})$  and  $u_\mu(\vec{q})$  are momentum and displacement operators, respectively for phonon modes in wave vector  $\vec{q}$  and polarization  $\mu$ . Strain field  $e_{ij}^{(s)}$  is defined the same as Eq.(3.2),  $e_{ij}^{(s)} = \frac{1}{2} \left( \partial u_i^{(s)} / \partial x_j + \partial u_j^{(s)} / \partial x_i \right)$ . The relation between displacement operator  $\vec{u}^{(s)}$  and  $\vec{u}_\mu(\vec{q})$  is set up by Fourier transformation:

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

where  $\vec{e}_\mu(\vec{q})$  is the unit vector representing the direction of vibrations,  $N$  is the number density of unit block, and by definition we automatically get  $Nm = \rho$ . For longitudinal mode  $\mu = l$ ,  $e_{li}(\vec{q}) = q_i/q$ , whereas for transverse modes  $t_1$  and  $t_2$ , we have,

$$\begin{aligned} \vec{e}_{t_1}(\vec{q}) \cdot \vec{q} = \vec{e}_{t_2}(\vec{q}) \cdot \vec{q} = \vec{e}_{t_1}(\vec{q}) \cdot \vec{e}_{t_1}(\vec{q}) = 0 \\ \sum_{\mu=t_1, t_2} e_{\mu i}(\vec{q}) e_{\mu j}(\vec{q}) = \delta_{ij} - \frac{q_i q_j}{q^2} \end{aligned} \quad (\text{A.4})$$

the strain field is therefore written as  $e_{ij}^{(s)} = \frac{1}{2\sqrt{N}} \sum_{\vec{q}\mu} i u_\mu(\vec{q}) e^{i\vec{q}\cdot\vec{x}_s} [q_j e_{\mu i}(\vec{q}) + q_i e_{\mu j}(\vec{q})]$ . Since for an arbitrary function  $f(\vec{q})$  we always have the following relation,  $\sum_{\vec{q}} f(\vec{q}) = \sum_{\vec{q}} \frac{1}{2} [f(\vec{q}) + f(-\vec{q})]$ , and the displacement  $u_i(\vec{x})$  is real, i.e.,  $u_i(\vec{x}) = u_i^*(\vec{x})$ , we have  $u_{\mu i}(\vec{q}) = u_{\mu i}^*(-\vec{q})$ . With these properties of  $u_\mu(\vec{q})$  operators we can rewrite the stress-strain coupling term as follows,

$$\sum_s \sum_{ij} e_{ij}^{(s)} \hat{T}_{ij}^{(s)} = \frac{1}{4\sqrt{N}} \sum_{ij} \sum_s \sum_{\vec{q}\mu} \left[ (i u_\mu(\vec{q}) e^{i\vec{q}\cdot\vec{x}_s}) + (i u_\mu(\vec{q}) e^{i\vec{q}\cdot\vec{x}_s})^* \right] (q_j e_{\mu i}(\vec{q}) + q_i e_{\mu j}(\vec{q})) \hat{T}_{ij}^{(s)} \quad (\text{A.5})$$

Because the stress-strain coupling term is linear in displacement operators  $u_\mu(\vec{q})$ , we can absorb it into terms quadratic in  $u_\mu(\vec{q})$ , i.e., the quadratic displacement term of phonon Hamiltonian, by completing the square.

An extra term comes out as follows:

$$\hat{H} = \sum_{\vec{q}\mu} \left( \frac{|p_\mu(\vec{q})|^2}{2m} + \frac{m\omega_{\vec{q}\mu}^2}{2} |u_\mu(\vec{q}) - u_\mu^{(0)}(\vec{q})|^2 - \frac{m\omega_{\vec{q}\mu}^2}{2} |u_\mu^{(0)}(\vec{q})|^2 \right) + \hat{H}^{\text{non}} \quad (\text{A.6})$$

where the ‘‘equilibrium position’’  $u_\mu^{(0)}(\vec{q})$  is

$$u_\mu^{(0)}(\vec{q}) = \frac{i}{2\sqrt{N}m\omega_{\vec{q}\mu}^2} \sum_{ij} \sum_s \left[ q_j e_{\mu i}(\vec{q}) + q_i e_{\mu j}(\vec{q}) \right] \hat{T}_{ij}^{(s)} e^{-i\vec{q}\cdot\vec{x}_s} \quad (\text{A.7})$$

The extra term left out after completing the square is the effective interaction between non-elastic stress tensors. It can be rewritten into two parts, the first part represents non-elastic stress-stress interaction within the same block, while the second part represents the interaction between different blocks:

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



We denote the second term in Eq.(A.8) as  $\hat{V}$ , non-elastic stress-stress interaction. Applying the properties of unit vector for longitudinal and transverse phonons, it is further simplified as

$$\begin{aligned}\hat{V} &= \frac{1}{2Nm} \left( \frac{1}{c_t^2} - \frac{1}{c_l^2} \right) \sum_{s \neq s'} \sum_{ijkl} \sum_{\vec{q}} \left( \frac{q_i q_j q_k q_l}{q^4} \right) \cos(\vec{q} \cdot \vec{x}_{ss'}) T_{ij}^{(s)} T_{kl}^{(s')} \\ &\quad - \frac{1}{8Nm} \frac{1}{c_t^2} \sum_{s \neq s'} \sum_{ijkl} \sum_{\vec{q}} \left( \frac{q_j q_l \delta_{ik} + q_j q_k \delta_{il} + q_i q_l \delta_{jk} + q_i q_k \delta_{jl}}{q^2} \right) \cos(\vec{q} \cdot \vec{x}_{ss'}) T_{ij}^{(s)} T_{kl}^{(s')}\end{aligned}\quad (\text{A.9})$$

where  $\vec{x}_{ss'} = \vec{x}_s - \vec{x}_{s'}$ . If we assume the inter-atomic distance is much smaller than phonon wavelength (long wavelength limit), we can use integral to replace summation over momentum  $\vec{q}$ . For convenience of discussion we write  $\hat{V}$  into two parts,  $\hat{V}^{(1)}$  and  $\hat{V}^{(2)}$ :

$$\begin{aligned}\hat{V}^{(1)} &= \frac{a^3}{2Nm} \left( \frac{1}{c_t^2} - \frac{1}{c_l^2} \right) \sum_{s \neq s'} \sum_{ijkl} \left\{ \int \frac{d^3 q}{(2\pi)^3} \left( \frac{q_i q_j q_k q_l}{q^4} \right) \cos(\vec{q} \cdot \vec{x}_{ss'}) \right\} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \\ \hat{V}^{(2)} &= -\frac{a^3}{8Nm} \frac{1}{c_t^2} \sum_{s \neq s'} \sum_{ijkl} \left\{ \int \frac{d^3 q}{(2\pi)^3} \left( \frac{q_j q_l \delta_{ik} + q_j q_k \delta_{il} + q_i q_l \delta_{jk} + q_i q_k \delta_{jl}}{q^2} \right) \cos(\vec{q} \cdot \vec{x}_{ss'}) \right\} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}\end{aligned}\quad (\text{A.10})$$

In the above two equations, we need to evaluate the following two integrals:

$$\begin{aligned}f_{ijkl}^{(1)} &= \int \frac{d^3 q}{(2\pi)^3} \frac{q_i q_j q_k q_l}{q^4} \cos(\vec{q} \cdot \vec{x}) \\ f_{jl}^{(2)} &= \int \frac{d^3 q}{(2\pi)^3} \frac{q_j q_l}{q^2} \cos(\vec{q} \cdot \vec{x})\end{aligned}\quad (\text{A.11})$$

Let us introduce a new parameter  $\lambda$  and take the limit  $\lambda \rightarrow 0$  eventually

$$\begin{aligned}f_{ijkl}^{(1)}(\lambda) &= \left( \frac{\partial}{\partial x_i} \frac{\partial}{\partial x_j} \frac{\partial}{\partial x_k} \frac{\partial}{\partial x_l} \right) \int \frac{d^3 q}{(2\pi)^3} \frac{1}{(q^2 + \lambda^2)^2} \frac{1}{2} (e^{i\vec{q} \cdot \vec{x}} + e^{-i\vec{q} \cdot \vec{x}}) \\ f_{jl}^{(2)}(\lambda) &= -\left( \frac{\partial}{\partial x_j} \frac{\partial}{\partial x_l} \right) \int \frac{d^3 q}{(2\pi)^3} \frac{1}{(q^2 + \lambda^2)} \frac{1}{2} (e^{i\vec{q} \cdot \vec{x}} + e^{-i\vec{q} \cdot \vec{x}})\end{aligned}\quad (\text{A.12})$$

Using contour integral, and choose the pole at  $q = -i\lambda$ , we have,

$$\begin{aligned}f_{ijkl}^{(1)}(\lambda) &= \left( \frac{\partial}{\partial x_i} \frac{\partial}{\partial x_j} \frac{\partial}{\partial x_k} \frac{\partial}{\partial x_l} \right) \frac{1}{8\pi\lambda} e^{-\lambda x} \\ f_{jl}^{(2)}(\lambda) &= -\left( \frac{\partial}{\partial x_j} \frac{\partial}{\partial x_l} \right) \frac{1}{4\pi x} e^{-\lambda x}\end{aligned}\quad (\text{A.13})$$

Finally, take the derivatives and we obtain,

$$\begin{aligned}
\lim_{\lambda \rightarrow 0} f_{ijkl}^{(1)}(\lambda) &= \frac{1}{8\pi x^3} \left\{ (\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{jk}\delta_{il}) \right. \\
&\quad \left. - 3(n_i n_j \delta_{kl} + n_i n_k \delta_{jl} + n_i n_l \delta_{jk} + n_j n_k \delta_{il} + n_j n_l \delta_{ik} + n_k n_l \delta_{ij}) + 15n_i n_j n_k n_l \right\} \\
\lim_{\lambda \rightarrow 0} f_{jl}^{(2)}(\lambda) &= \frac{1}{4\pi x^3} (\delta_{jl} - 3n_j n_l)
\end{aligned} \tag{A.14}$$

Finally, plugging the above results of integrals, we eventually get our non-elastic stress-stress interaction coefficient  $\Lambda_{ijkl}^{(ss')}$

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

Compare  $\Lambda_{ijkl}^{(ss')}$  in Eq.(A.15) and  $\left(\Lambda_{ijkl}^{(ss')}\right)_{\text{Joffrin}}$  in Eq.(A.1), there are 4 differences between our result and Joffrin and Levelut's result:

- (1). The first term without  $\alpha$  in  $\Lambda_{ijkl}^{(ss')}$ , is  $\frac{1}{4} \{ (\delta_{jl} - 3n_j n_l) \delta_{ik} + (\delta_{jk} - 3n_j n_k) \delta_{il} + (\delta_{ik} - 3n_i n_k) \delta_{jl} + (\delta_{il} - 3n_i n_l) \delta_{jk} \}$ , while it is  $(\delta_{jl} - 3n_j n_l) \delta_{ik}$  in  $\left(\Lambda_{ijkl}^{(ss')}\right)_{\text{Joffrin}}$ . This difference is fine, because in Joffrin and Levelut's calculation their strain tensor  $e_{ij}$  is defined as  $\partial u_i / \partial x_j$ , while ours is symmetrized:  $(\partial u_i / \partial x_j + \partial u_j / \partial x_i) / 2$ . Using the definition  $e_{ij} = \partial u_i / \partial x_j$  at the start of our calculation will give the same unpermuted result  $(\delta_{jl} - 3n_j n_l) \delta_{ik}$ ;
- (2). Our  $\Lambda_{ijkl}^{(ss')}$  is smaller by a total factor of 1/2 compared to  $\left(\Lambda_{ijkl}^{(ss')}\right)_{\text{Joffrin}}$ ;
- (3). We do not have the negative sign in the term not multiplied by  $\alpha$ , while Joffrin and Levelut's term has:  $-(\delta_{jl} - 3n_j n_l) \delta_{ik}$ ;
- (4). The second term which is multiplied by  $\alpha$  has an extra factor of 1/2 in our  $\Lambda_{ijkl}^{(ss')}$  compared to  $\left(\Lambda_{ijkl}^{(ss')}\right)_{\text{Joffrin}}$ .

## Appendix B

# Derivations of Renormalization Equation of Non-elastic Stress-Stress Susceptibility

We have super block susceptibility given as follows,

$$\begin{aligned}
\chi_{ijkl}^{\text{super}}(\omega) &= \frac{1}{L'^3} \frac{\beta}{1 - i\omega\tau^*} \left( \sum_{n^*m^*} \frac{e^{-\beta(E_n^* + E_m^*)}}{\mathcal{Z}^{*2}} \langle n^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle m^* | \hat{T}_{kl}^{\text{super}} | m^* \rangle \right. \\
&\quad \left. - \sum_{n^*} \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \langle n^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle n^* | \hat{T}_{kl}^{\text{super}} | n^* \rangle \right) \\
&+ \frac{1}{L'^3} \frac{2}{\hbar} \sum_{n^*l^*} \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \langle l^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle n^* | \hat{T}_{kl}^{\text{super}} | l^* \rangle \frac{\omega_l^* - \omega_n^*}{(\omega + i\eta)^2 - (\omega_l^* - \omega_n^*)^2}
\end{aligned} \tag{B.1}$$

where  $L' = N_0L$ . We want to find the relation between super block susceptibility  $\chi_{ijkl}^{\text{super}}(\omega)$  and single block susceptibility  $\chi_{ijkl}(\omega)$ . Please note: in the following calculations (renormalization procedures) we are only interested in the first and second orders of susceptibility, which means we only take 2nd and 4th order in  $\hat{T}_{ij}$  into account. We will drop terms in 3rd order of stress tensor  $\hat{T}_{ij}$ .

We treat  $\hat{V}$  as perturbation. By using time-independent perturbation theory, we obtain

$$|n^*\rangle = |n\rangle + \sum_p \frac{\langle p | \hat{V} | n \rangle}{E_n - E_p} |p\rangle + \dots \quad E_n^* = E_n + \langle n | \hat{V} | n \rangle + \sum_{p \neq n} \frac{|\langle p | \hat{V} | n \rangle|^2}{E_n - E_p} + \dots \tag{B.2}$$

Therefore we have to expand the distribution function and probability function up to the second order in  $\hat{V}$ :

$$\begin{aligned}
e^{-\beta E_n^*} &= e^{-\beta E_n} \left( 1 - \beta \langle n | \hat{V} | n \rangle - \beta \sum_{p \neq n} \frac{|\langle p | \hat{V} | n \rangle|^2}{E_n - E_p} + \dots \right) \\
\mathcal{Z} &= \sum_l e^{-\beta E_l} \left( 1 - \beta \langle l | \hat{V} | l \rangle - \beta \sum_{p \neq l} \frac{|\langle p | \hat{V} | l \rangle|^2}{E_l - E_p} + \dots \right)
\end{aligned} \tag{B.3}$$

The following definitions are much more useful in details of calculations:

$$\begin{aligned}
\chi_{ijkl}^{\text{rel}(1)} &= \frac{1}{L^3} \beta \sum_{nm} P_n P_m \langle n | \hat{T}_{ij} | n \rangle \langle m | \hat{T}_{kl} | m \rangle \\
\chi_{ijkl}^{\text{rel}(2)} &= \frac{1}{L^3} \beta \sum_n P_n \langle n | \hat{T}_{ij} | n \rangle \langle n | \hat{T}_{kl} | n \rangle \\
\chi_{ijkl}^{\text{res}}(\omega + i\eta) &= \frac{1}{L^3} \frac{1}{\hbar} \sum_{nl} (P_n - P_l) \frac{\langle n | \hat{T}_{ij} | l \rangle \langle l | \hat{T}_{kl} | n \rangle}{\omega + \omega_{nl} + i\eta}
\end{aligned} \tag{B.4}$$

hence

$$\chi_{ijkl}(\omega) = \frac{1}{1 - i\omega\tau} \left( \chi_{ijkl}^{\text{rel}(1)}(\omega) - \chi_{ijkl}^{\text{rel}(2)}(\omega) \right) + \chi_{ijkl}^{\text{res}}(\omega + i\eta) \tag{B.5}$$

In the following of this appendix we want to expand three parts of super block non-elastic susceptibility,  $\chi_{ijkl}^{\text{super rel}(1)}$ ,  $\chi_{ijkl}^{\text{super rel}(2)}$  and  $\chi_{ijkl}^{\text{super res}}(\omega + i\eta)$  up to the first order of interaction  $\hat{V}$  (i.e., the second order of unit block susceptibility). From Eq.(3.16) we know there is an extra term in super block stress tensor generated by the strain field dependence of coefficient  $\Lambda_{abcd}^{(ss')}(e)$ . Let's denote  $\sum_s e^{i\vec{k}\cdot\vec{x}_s} \hat{T}_{ij}^{(s)} = \hat{T}_{ij}$ , so we have  $\hat{T}_{ij}^{\text{super}} = \hat{T}_{ij} + \sum_{s \neq s'} \sum_{abcd} e^{i\vec{k}\cdot\frac{\vec{x}_s + \vec{x}_{s'}}{2}} \frac{\delta \Lambda_{abcd}^{(ss')}(e)}{\delta e_{ij}} \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')}$ . We will discuss higher order expansions from the extra term in stress tensor  $\sum_{s \neq s'} \sum_{abcd} e^{i\vec{k}\cdot\frac{\vec{x}_s + \vec{x}_{s'}}{2}} \frac{\delta \Lambda_{abcd}^{(ss')}(e)}{\delta e_{ij}} \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')}$  in the last section of this appendix. Currently we consider higher order expansions of super block susceptibility with stress tensor  $\hat{T}_{ij}$ .

## B.1 Expansion details for $\chi_{ijkl}^{\text{super rel}(1)}$

$$\begin{aligned}
& \frac{\beta}{(N_0 L)^3} \sum_{n^* m^*} \frac{e^{-\beta(E_{n^*} + E_{m^*})}}{\mathcal{Z}^{*2}} \langle n^* | \hat{T}_{ij,cc} | n^* \rangle \langle m^* | \hat{T}_{kl} | m^* \rangle \\
&= \frac{\beta}{(N_0 L)^3} \sum_{nm} \frac{e^{-\beta(E_n + E_m)}}{\mathcal{Z}^2} \langle n | \hat{T}_{ij,cc} | n \rangle \langle m | \hat{T}_{kl} | m \rangle \\
\text{term(1)} &+ \frac{\beta}{(N_0 L)^3} \sum_{nm} \frac{e^{-\beta(E_n + E_m)} (-\beta \delta E_n - \beta \delta E_m)}{\mathcal{Z}^2} \langle n | \hat{T}_{ij,cc} | n \rangle \langle m | \hat{T}_{kl} | m \rangle \\
\text{term(2)} &+ \frac{\beta}{(N_0 L)^3} \sum_{nm} \frac{e^{-\beta(E_n + E_m)} (-2\delta \mathcal{Z})}{\mathcal{Z}^3} \langle n | \hat{T}_{ij,cc} | n \rangle \langle m | \hat{T}_{kl} | m \rangle \\
\text{term(3)} &+ \frac{\beta}{(N_0 L)^3} \sum_{nm} \frac{e^{-\beta(E_n + E_m)}}{\mathcal{Z}^2} \left[ (\delta \langle n |) \hat{T}_{ij,cc} | n \rangle \langle m | \hat{T}_{kl} \rangle + \langle n | \hat{T}_{ij,cc} (\delta | n \rangle) \langle m | \hat{T}_{kl} \rangle \right. \\
&\quad \left. + \langle n | \hat{T}_{ij,cc} | n \rangle (\delta \langle m |) \hat{T}_{kl} \rangle + \langle n | \hat{T}_{ij,cc} | n \rangle \langle m | \hat{T}_{kl} (\delta | m \rangle) \right] \tag{B.6}
\end{aligned}$$

where  $\hat{T}_{ij,cc}$  is the complex conjugate of  $\hat{T}_{ij}$ .  $\delta\mathcal{Z}$  and  $\delta E_n$  represents first and second order expansions with respect to many body interaction  $\hat{V}$ . Now we begin to calculate every expansions in the above result.

Expansion for term(1):

$$\begin{aligned}
& -\frac{\beta^2}{(N_0L)^3} \sum_{nm} \frac{e^{-\beta(E_n+E_m)}(\delta E_n + \delta E_m)}{\mathcal{Z}^2} \langle n|\hat{T}_{ij,cc}|n\rangle \langle m|\hat{T}_{kl}|m\rangle \\
= & -\frac{\beta^2}{(N_0L)^3} \sum_{nm} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \left( \langle n|\hat{V}|n\rangle + \langle m|\hat{V}|m\rangle \right) \langle n|\hat{T}_{ij,cc}|n\rangle \langle m|\hat{T}_{kl}|m\rangle \\
= & -\frac{\beta^2}{(N_0L)^3} \sum_{nm} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
& \left( \langle n|\hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')}|n\rangle + \langle m|\hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')}|m\rangle \right) \langle n|\hat{T}_{ij}^{(s)}|n\rangle \langle m|\hat{T}_{kl}^{(s')}|m\rangle \\
= & -\frac{\beta^2}{(N_0L)^3} \sum_{nm} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
& \left( \langle n|\hat{T}_{ab}^{(u)} \sum_l |l\rangle \langle l|\hat{T}_{cd}^{(u')}|n\rangle + \langle m|\hat{T}_{ab}^{(u)} \sum_l |l\rangle \langle l|\hat{T}_{cd}^{(u')}|m\rangle \right) \langle n|\hat{T}_{ij}^{(s)}|n\rangle \langle m|\hat{T}_{kl}^{(s')}|m\rangle
\end{aligned}$$

We only defined the relaxation susceptibility, which is the product between diagonal matrix elements of  $\hat{T}_{ij}$ , and the resonance susceptibility which is the product of off-diagonal matrix elements of  $\hat{T}_{ij}$ . We have never defined the product between diagonal and off-diagonal matrix elements of  $\hat{T}_{ij}$ . The reason is if we average over spacial coordinate such kind of diagonal-off-diagonal matrix element product will vanish for the random distribution of matrix element values if glass. In other words, there is no specific relation between diagonal and off-diagonal matrix elements. In addition, the diagonal matrix element  $\langle n|\hat{T}_{ij}^{(s)}|n\rangle \propto \delta\langle \hat{H}^{\text{non}}\rangle/\delta e_{ij}$  which is the ‘‘total’’ stress tensor minus elastic stress tensor. It is highly plausible that the non-elastic stress tensor expectation value tends to vanish for large enough block of glass. Because we want to pair the matrix elements in the above equation, the only choices for quantum number  $l$  is  $l = n, m$ . Because  $u \neq u'$  for  $\Lambda_{ijkl}^{(uu')}$ , we also have to pair  $u$  with  $s$  or  $s'$ . So we have two choices,  $u = s$ , or  $u = s'$ .

$$\begin{aligned}
= & -\frac{\beta^2}{(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{n^{(s)}n^{(s')}m^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')}+E_m^{(s')})}}{\mathcal{Z}^{(s)}\mathcal{Z}^{(s')2}} \Lambda_{abcd}^{(ss')} e^{-ik(x_s-x'_s)} \\
& \left\{ \langle n^{(s)}|\hat{T}_{cd}^{(s)}|n^{(s)}\rangle \langle n^{(s)}|\hat{T}_{ij}^{(s)}|n^{(s)}\rangle \langle m^{(s')}|\hat{T}_{kl}^{(s')}|m^{(s')}\rangle \langle n^{(s')}|\hat{T}_{ab}^{(s')}|n^{(s')}\rangle \right. \\
& \left. + \langle n^{(s)}|\hat{T}_{ij}^{(s)}|n^{(s)}\rangle \langle m^{(s)}|\hat{T}_{ab}^{(s)}|m^{(s)}\rangle \langle m^{(s')}|\hat{T}_{cd}^{(s')}|m^{(s')}\rangle \langle m^{(s')}|\hat{T}_{kl}^{(s')}|m^{(s')}\rangle \right\} \\
= & -\frac{L^6}{(N_0L)^3} \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-ik(x_s-x'_s)} \left( \chi_{cdij}^{\text{rel}(2)} \chi_{abkl}^{\text{rel}(1)} + \chi_{cdkl}^{\text{rel}(2)} \chi_{abij}^{\text{rel}(1)} \right) \tag{B.7}
\end{aligned}$$

Expansion for term(2):

$$\begin{aligned}
& -\frac{2\beta}{(N_0L)^3} \sum_{nm} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^3} \delta\mathcal{Z} \langle n|\hat{T}_{ij,cc}|n\rangle \langle m|\hat{T}_{kl}|m\rangle \\
&= \frac{2\beta^2}{(N_0L)^3} \sum_{lmn} \frac{e^{-\beta(E_n+E_m+E_l)}}{\mathcal{Z}^3} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik\cdot(x_s-x'_s)} \langle l|\hat{T}_{ab}^{(u)}\hat{T}_{cd}^{(u')}|l\rangle \langle n|\hat{T}_{ij}^{(s)}|n\rangle \langle m|\hat{T}_{kl}^{(s')}|m\rangle \\
&= \frac{2\beta^2}{(N_0L)^3} \sum_{lmn} \frac{e^{-\beta(E_n+E_m+E_l)}}{\mathcal{Z}^3} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik\cdot(x_s-x'_s)} \\
&\quad \langle l|\hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k|\hat{T}_{cd}^{(u')}|l\rangle \langle n|\hat{T}_{ij}^{(s)}|n\rangle \langle m|\hat{T}_{kl}^{(s')}|m\rangle \\
&= \frac{2\beta^2}{(N_0L)^3} \sum_{l^{(s)}l^{(s')}m^{(s')}n^{(s)}} \frac{e^{-\beta(E_n^{(s)}+E_m^{(s')}+E_l^{(s)}+E_l^{(s')})}}{\mathcal{Z}^{(s)2}\mathcal{Z}^{(s')2}} \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-ik\cdot(x_s-x'_s)} \\
&\quad \langle l^{(s')}|\hat{T}_{cd}^{(s')}|l^{(s')}\rangle \langle m^{(s')}|\hat{T}_{kl}^{(s')}|m^{(s')}\rangle \langle n^{(s)}|\hat{T}_{ij}^{(s)}|n^{(s)}\rangle \langle l^{(s)}|\hat{T}_{ab}^{(s)}|l^{(s)}\rangle \\
&= \frac{2L^6}{(N_0L)^3} \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{ik\cdot(x_s-x'_s)} \chi_{cdij}^{\text{rel}(1)} \chi_{abkl}^{\text{rel}(1)} \tag{B.8}
\end{aligned}$$

where in the above calculation we need to pair diagonal matrix elements  $\langle n|\hat{T}_{ij}^{(s)}|n\rangle$  and  $\langle m|\hat{T}_{kl}^{(s')}|m\rangle$ , so the choice of  $k$  have to be  $k = l$ , to make the matrix element  $\langle l|\hat{T}_{ab}^{(u)}|k\rangle$  diagonal. Then it could be paired to  $\langle n|\hat{T}_{ij}^{(s)}|n\rangle$  or  $\langle m|\hat{T}_{kl}^{(s')}|m\rangle$ . The reason it must be pair to  $\langle n|\hat{T}_{ij}^{(s)}|n\rangle$  or  $\langle m|\hat{T}_{kl}^{(s')}|m\rangle$  is because from the coefficient  $\Lambda_{ijkl}^{(uu')}$ ,  $n \neq u'$  so the matrix elements  $\langle l|\hat{T}_{ab}^{(u)}|k\rangle \langle k|\hat{T}_{cd}^{(u')}|l\rangle$  cannot be paired. Finally, we have two choices for  $u, u'$ :  $u = s$  and  $u' = s'$ , or  $u = s'$  and  $u' = s$ .

Expansion for term(3):

$$\begin{aligned}
& \frac{\beta}{(N_0 L)^3} \sum_{nm} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \left[ (\delta\langle n|) \hat{T}_{ij,cc} |n\rangle \langle m| \hat{T}_{kl} |m\rangle + \langle n| \hat{T}_{ij,cc} (\delta|n\rangle) \langle m| \hat{T}_{kl} |m\rangle \right. \\
& \quad \left. + \langle n| \hat{T}_{ij,cc} |n\rangle (\delta\langle m|) \hat{T}_{kl} |m\rangle + \langle n| \hat{T}_{ij,cc} |n\rangle \langle m| \hat{T}_{kl} (\delta|m\rangle) \right] \\
&= \frac{\beta}{(N_0 L)^3} \sum_{lmn} \sum_{abcd} \sum_{uu'} \sum_{ss'} \frac{1}{E_n - E_l} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
& \quad \left( \langle n| \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} |l\rangle \langle l| \hat{T}_{ij}^{(s)} |n\rangle \langle m| \hat{T}_{kl}^{(s')} |m\rangle + \langle n| \hat{T}_{ij}^{(s)} |l\rangle \langle l| \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} |n\rangle \langle m| \hat{T}_{kl}^{(s')} |m\rangle \right) \\
&+ \frac{\beta}{(N_0 L)^3} \sum_{lmn} \sum_{abcd} \sum_{uu'} \sum_{ss'} \frac{1}{E_m - E_l} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
& \quad \left( \langle n| \hat{T}_{ij}^{(s)} |n\rangle \langle m| \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} |l\rangle \langle l| \hat{T}_{kl}^{(s')} |m\rangle + \langle n| \hat{T}_{ij}^{(s)} |n\rangle \langle m| \hat{T}_{kl}^{(s')} |l\rangle \langle l| \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} |m\rangle \right) \\
&= \frac{\beta}{(N_0 L)^3} \sum_{lmn} \sum_{abcd} \sum_{uu'} \sum_{ss'} \frac{1}{E_n - E_l} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
& \quad \left( \langle n| \hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k| \hat{T}_{cd}^{(u')} |l\rangle \langle l| \hat{T}_{ij}^{(s)} |n\rangle \langle m| \hat{T}_{kl}^{(s')} |m\rangle + \langle n| \hat{T}_{ij}^{(s)} |l\rangle \langle l| \hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k| \hat{T}_{cd}^{(u')} |n\rangle \langle m| \hat{T}_{kl}^{(s')} |m\rangle \right) \\
&+ \frac{\beta}{(N_0 L)^3} \sum_{lmn} \sum_{abcd} \sum_{uu'} \sum_{ss'} \frac{1}{E_m - E_l} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
& \quad \left( \langle n| \hat{T}_{ij}^{(s)} |n\rangle \langle m| \hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k| \hat{T}_{cd}^{(u')} |l\rangle \langle l| \hat{T}_{kl}^{(s')} |m\rangle + \langle n| \hat{T}_{ij}^{(s)} |n\rangle \langle m| \hat{T}_{kl}^{(s')} |l\rangle \langle l| \hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k| \hat{T}_{cd}^{(u')} |m\rangle \right) \\
&= \frac{2\beta}{(N_0 L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s)} m^{(s')} n^{(s)} n^{(s')}} \frac{e^{-\beta(E_n^{(s)} + E_n^{(s')} + E_m^{(s')})}}{\mathcal{Z}^{(s)} \mathcal{Z}^{(s')2}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \\
& \quad \langle m^{(s')} | \hat{T}_{kl}^{(s')} | m^{(s')} \rangle \langle n^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle \frac{\langle l^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ab}^{(s)} | l^{(s)} \rangle}{E_n^{(s)} - E_l^{(s)}} \\
&+ \frac{2\beta}{(N_0 L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')} m^{(s')} n^{(s)} m^{(s)}} \frac{e^{-\beta(E_n^{(s)} + E_m^{(s)} + E_m^{(s')})}}{\mathcal{Z}^{(s)2} \mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \\
& \quad \langle n^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ab}^{(s)} | m^{(s)} \rangle \frac{\langle m^{(s')} | \hat{T}_{kl}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{cd}^{(s')} | m^{(s')} \rangle}{E_m^{(s')} - E_l^{(s')}} \\
&= \frac{L^6}{(N_0 L)^3} \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \left( \chi_{cdkl}^{\text{rel}(1)} \chi_{abij}^{\text{res}}(0) + \chi_{abij}^{\text{rel}(1)} \chi_{cdkl}(0) \right) \tag{B.9}
\end{aligned}$$

where in the above calculations we insert the identity between  $\hat{T}_{ab}^{(u)}$  and  $\hat{T}_{cd}^{(u')}$ :  $\sum_k |k\rangle \langle k|$ . Because of the coefficient  $\lambda_{abcd}^{(uu')}$  we know  $u \neq u'$ . Therefore if we want to make matrix elements in pairs, the only choices are  $u = s$  and  $u' = s'$  or  $u = s'$  and  $u' = s$ . Since the matrix element for the  $s$ -th block stress tensor is diagonal, while for the  $s'$ -th block it is off-diagonal, we need to pair on diagonal matrix element to the  $s$ -th block stress tensor, and pair a off-diagonal matrix element to the  $s'$ -th block stress tensor. For example, we could pair  $u = s$  and  $u' = s'$ , another case is similar. Therefore the wavefunction  $|m\rangle$  must equal to  $|k\rangle$ :

$\delta_{mk}$  for the diagonal matrix element pairing, on the other hand, the wave function  $|k\rangle$  must equal to  $|m\rangle$  for the off-diagonal matrix element pairing.

## B.2 Expansion details for $\chi_{ijkl}^{\text{super rel}(2)}$

$$\begin{aligned}
& -\frac{\beta}{(N_0L)^3} \sum_{n^*} \frac{e^{-\beta E_{n^*}}}{\mathcal{Z}^*} \langle n^* | \hat{T}_{ij,cc} | n^* \rangle \langle n^* | \hat{T}_{kl} | n^* \rangle \\
&= -\frac{\beta}{(N_0L)^3} \sum_n \frac{e^{-\beta E_n} (1 - \beta E_n)}{\mathcal{Z} + \delta \mathcal{Z}} (\langle n | + \delta \langle n |) \hat{T}_{ij,cc} (|n\rangle + \delta |n\rangle) (\langle n | + \delta \langle n |) \hat{T}_{kl} (|n\rangle + \delta |n\rangle) \\
&= -\frac{\beta}{(N_0L)^3} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \langle n | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | n \rangle \\
\text{term(4)} \quad & -\frac{\beta}{(N_0L)^3} \sum_n \frac{e^{-\beta E_n} (-\beta \delta E_n)}{\mathcal{Z}} \langle n | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | n \rangle \\
\text{term(5)} \quad & -\frac{\beta}{(N_0L)^3} \sum_n \frac{-e^{-\beta E_n}}{\mathcal{Z}^2} \delta \mathcal{Z} \langle n | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | n \rangle \\
\text{term(6)} \quad & -\frac{\beta}{(N_0L)^3} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \left[ (\delta \langle n |) \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | n \rangle + \langle n | \hat{T}_{ij,cc} (\delta |n\rangle) \langle n | \hat{T}_{kl} | n \rangle \right. \\
& \quad \left. + \langle n | \hat{T}_{ij,cc} | n \rangle (\delta \langle n |) \hat{T}_{kl} | n \rangle + \langle n | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} (\delta |n\rangle) \right] \tag{B.10}
\end{aligned}$$

Expansion for term(4):

$$\begin{aligned}
& \frac{\beta^2}{(N_0L)^3} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \delta E_n \langle n | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | n \rangle \\
&= \frac{\beta^2}{(N_0L)^3} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \langle n | \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | n \rangle \langle n | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | n \rangle \\
&= \frac{\beta^2}{(N_0L)^3} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \langle n | \hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k| \hat{T}_{cd}^{(u')} | n \rangle \langle n | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | n \rangle \\
&= \frac{\beta^2}{(N_0L)^3} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \langle n^{(s)} | \hat{T}_{ab}^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \\
& \quad \langle n^{(s')} | \hat{T}_{kl}^{(s')} | n^{(s')} \rangle \langle n^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle \\
&= \frac{L^6}{(N_0L)^3} \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \chi_{abij}^{\text{rel}(2)} \chi_{cdkl}^{\text{rel}(2)} \tag{B.11}
\end{aligned}$$

where in the above calculation we insert the identity  $\sum_k |k\rangle \langle k|$ . Because of the coefficient  $\Lambda_{abcd}^{(uu')}$  we have  $u \neq u'$  so if we want to make the matrix element come in pairs, we need to pair  $u = s$  and  $u' = s'$  or  $u = s'$  and  $u' = s$ . Since the matrix elements for stress tensor  $\hat{T}^{(s)}$  and  $\hat{T}^{(s')}$  are diagonal, we must pair diagonal matrix elements from  $\hat{T}^{(u)}$  and  $\hat{T}^{(u')}$  onto them. Therefore the only choice for  $k$  is  $k = n$ .



Expansion for term(5):

$$\begin{aligned}
& \frac{\beta}{(N_0 L)^3} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}^2} \delta \mathcal{Z} \langle n | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | n \rangle \\
= & -\frac{\beta^2}{(N_0 L)^3} \sum_{nl} \frac{e^{-\beta(E_n+E_l)}}{\mathcal{Z}^2} \langle l | \hat{V} | l \rangle \langle n | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | n \rangle \\
= & -\frac{\beta^2}{(N_0 L)^3} \sum_{nl} \frac{e^{-\beta(E_n+E_l)}}{\mathcal{Z}^2} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \langle l | \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | l \rangle \langle n | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | n \rangle \\
= & -\frac{\beta^2}{(N_0 L)^3} \sum_{nl} \frac{e^{-\beta(E_n+E_l)}}{\mathcal{Z}^2} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
& \langle l | \hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k| \hat{T}_{cd}^{(u')} | l \rangle \langle n | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | n \rangle \\
= & -\frac{\beta^2}{(N_0 L)^3} \sum_{lmn} \frac{e^{-\beta(E_n+E_l)}}{\mathcal{Z}^2} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
& \langle l | \hat{T}_{ab}^{(u)} | m \rangle \langle m | \hat{T}_{cd}^{(u')} | l \rangle \langle n | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | n \rangle \\
= & -\frac{\beta^2}{(N_0 L)^3} \sum_{ss'} \sum_{l^{(s)} l^{(s')} n^{(s)} n^{(s')}} \frac{e^{-\beta(E_n^{(s)} + E_n^{(s')} + E_l^{(s)} + E_l^{(s')})}}{\mathcal{Z}^{(s)2} \mathcal{Z}^{(s')2}} \sum_{abcd} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \\
& \langle l^{(s)} | \hat{T}_{cd}^{(s)} | l^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle l^{(s')} | \hat{T}_{ab}^{(s')} | l^{(s')} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | n^{(s')} \rangle \\
= & -\frac{L^6}{(N_0 L)^3} \sum_{ss'} \sum_{abcd} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \chi_{abij}^{\text{rel}(1)} \chi_{cdkl}^{\text{rel}(1)} \tag{B.12}
\end{aligned}$$

where in the above calculations we have inserted the identity  $\sum_k |k\rangle \langle k|$ . Because of the coefficient  $\Lambda_{abcd}^{(uu')}$  we have  $u \neq u'$ . So we have to pair the matrix element of  $\hat{T}^{(u)}$  with  $\hat{T}^{(s)}$  and  $\hat{T}^{(s')}$ , and the same for  $\hat{T}^{(u')}$ . Since the matrix elements for  $\hat{T}^{(s)}$  and  $\hat{T}^{(s')}$  are diagonal matrix elements, we also have to pair the diagonal matrix elements of  $\hat{T}^{(u)}$  and  $\hat{T}^{(u')}$ . Thus the only choice for  $k$  is  $k = l$ .

Expansion for term(6):

$$\begin{aligned}
& -\frac{\beta}{(N_0L)^3} \sum_{nm} \frac{1}{E_n - E_m} \frac{e^{-\beta E_n}}{\mathcal{Z}} \\
& \left[ \langle n|\hat{V}|m\rangle \langle m|\hat{T}_{ij,cc}|n\rangle \langle n|\hat{T}_{kl}|n\rangle + \langle n|\hat{T}_{ij,cc}|m\rangle \langle m|\hat{V}|n\rangle \langle n|\hat{T}_{kl}|n\rangle \right. \\
& \left. + \langle n|\hat{T}_{ij,cc}|n\rangle \langle n|\hat{V}|m\rangle \langle m|\hat{T}_{kl}|n\rangle + \langle n|\hat{T}_{ij,cc}|n\rangle \langle n|\hat{T}_{kl}|m\rangle \langle m|\hat{V}|n\rangle \right] \\
= & -\frac{\beta}{(N_0L)^3} \sum_{nml} \sum_{abcd} \sum_{uu'} \sum_{ss'} \frac{1}{E_n - E_m} \frac{e^{-\beta E_n}}{\mathcal{Z}} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
& \left\{ \langle n|\hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k|\hat{T}_{cd}^{(u')} |m\rangle \langle m|\hat{T}_{ij}^{(s)} |n\rangle \langle n|\hat{T}_{kl}^{(s')} |n\rangle + \langle n|\hat{T}_{ij}^{(s)} |m\rangle \langle m|\hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k|\hat{T}_{cd}^{(u')} |n\rangle \langle n|\hat{T}_{kl}^{(s')} |n\rangle \right. \\
& \left. + \langle n|\hat{T}_{ij}^{(s)} |n\rangle \langle n|\hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k|\hat{T}_{cd}^{(u')} |m\rangle \langle m|\hat{T}_{kl}^{(s')} |n\rangle + \langle n|\hat{T}_{ij}^{(s)} |n\rangle \langle n|\hat{T}_{kl}^{(s')} |m\rangle \langle m|\hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k|\hat{T}_{cd}^{(u')} |n\rangle \right\} \\
= & -\frac{\beta}{(N_0L)^3} \sum_{nml} \sum_{abcd} \sum_{uu'} \sum_{ss'} \frac{1}{E_n - E_m} \frac{e^{-\beta E_n}}{\mathcal{Z}} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
& \left\{ \langle n|\hat{T}_{ab}^{(u)} |l\rangle \langle l|\hat{T}_{cd}^{(u')} |m\rangle \langle m|\hat{T}_{ij}^{(s)} |n\rangle \langle n|\hat{T}_{kl}^{(s')} |n\rangle + \langle n|\hat{T}_{ij}^{(s)} |m\rangle \langle m|\hat{T}_{ab}^{(u)} |l\rangle \langle l|\hat{T}_{cd}^{(u')} |n\rangle \langle n|\hat{T}_{kl}^{(s')} |n\rangle \right. \\
& \left. + \langle n|\hat{T}_{ij}^{(s)} |n\rangle \langle n|\hat{T}_{ab}^{(u)} |l\rangle \langle l|\hat{T}_{cd}^{(u')} |m\rangle \langle m|\hat{T}_{kl}^{(s')} |n\rangle + \langle n|\hat{T}_{ij}^{(s)} |n\rangle \langle n|\hat{T}_{kl}^{(s')} |m\rangle \langle m|\hat{T}_{ab}^{(u)} |l\rangle \langle l|\hat{T}_{cd}^{(u')} |n\rangle \right\} \\
= & -\frac{\beta}{(N_0L)^3} \sum_{nml} \sum_{abcd} \sum_{ss'} \frac{1}{E_n - E_m} \frac{e^{-\beta E_n}}{\mathcal{Z}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \\
& \left\{ \langle n|\hat{T}_{ab}^{(s')} |l\rangle \langle l|\hat{T}_{cd}^{(s)} |m\rangle \langle m|\hat{T}_{ij}^{(s)} |n\rangle \langle n|\hat{T}_{kl}^{(s')} |n\rangle + \langle n|\hat{T}_{ij}^{(s)} |m\rangle \langle m|\hat{T}_{ab}^{(s')} |l\rangle \langle l|\hat{T}_{cd}^{(s)} |n\rangle \langle n|\hat{T}_{kl}^{(s')} |n\rangle \right. \\
& \left. + \langle n|\hat{T}_{ij}^{(s)} |n\rangle \langle n|\hat{T}_{ab}^{(s')} |l\rangle \langle l|\hat{T}_{cd}^{(s)} |m\rangle \langle m|\hat{T}_{kl}^{(s')} |n\rangle + \langle n|\hat{T}_{ij}^{(s)} |n\rangle \langle n|\hat{T}_{kl}^{(s')} |m\rangle \langle m|\hat{T}_{ab}^{(s')} |l\rangle \langle l|\hat{T}_{cd}^{(s)} |n\rangle \right\} \\
= & -\frac{2\beta}{(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{n^{(s)}n^{(s')}m^{(s)}} \frac{1}{E_n^{(s)} - E_m^{(s)}} \frac{e^{-\beta(E_n^{(s)} + E_n^{(s')})}}{\mathcal{Z}^{(s)}\mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \\
& \langle n^{(s')}|\hat{T}_{kl}^{(s')} |n^{(s')}\rangle \langle n^{(s')}|\hat{T}_{ab}^{(s)} |n^{(s')}\rangle \langle m^{(s)}|\hat{T}_{cd}^{(s)} |n^{(s)}\rangle \langle n^{(s)}|\hat{T}_{ij}^{(s)} |m^{(s)}\rangle \\
+ & -\frac{2\beta}{(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{n^{(s)}n^{(s')}m^{(s')}} \frac{1}{E_n^{(s')} - E_m^{(s')}} \frac{e^{-\beta(E_n^{(s)} + E_n^{(s')})}}{\mathcal{Z}^{(s)}\mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \\
& \langle n^{(s)}|\hat{T}_{ij}^{(s)} |n^{(s)}\rangle \langle n^{(s)}|\hat{T}_{ab}^{(s)} |n^{(s)}\rangle \langle n^{(s')}|\hat{T}_{cd}^{(s')} |m^{(s')}\rangle \langle m^{(s')}|\hat{T}_{kl}^{(s')} |n^{(s')}\rangle \\
= & -\frac{L^6}{(N_0L)^3} \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \left( \chi_{abkl}^{\text{rel}(2)} \chi_{cdij}^{\text{res}}(0) + \chi_{abij}^{\text{rel}(2)} \chi_{cdkl}^{\text{res}}(0) \right) \tag{B.13}
\end{aligned}$$

where in the above second step we insert the identity  $\sum_k |k\rangle \langle k|$ . Since the presence of coefficient  $\Lambda_{abcd}^{(uu')}$ ,  $u$  cannot equal to  $u'$ . So we need to pair stress tensors in  $u$ -th and  $u'$ -th block to the  $s$ -th and  $s'$ -th block stress tensors. The matrix element for the  $s$ -th and  $s'$ -th block stress tensor is one diagonal, one off-diagonal. We need to pair one diagonal matrix element and one off-diagonal matrix element for stress tensors at block position  $u$  and  $u'$ . For example, we consider  $u = s$  and  $u' = s'$ . The other case is similar. The term

$\langle n|\hat{T}_{ab}^{(u=s)}\sum_k|k\rangle\langle k|\hat{T}_{cd}^{(u'=s')}|m\rangle\langle m|\hat{T}_{ij}^{(s)}|n\rangle\langle n|\hat{T}_{kl}^{(s')}|n\rangle$  does not vanish only when  $\langle k|\hat{T}_{cd}^{(u'=s')}|m\rangle$  is diagonal, and it can be paired to  $\langle n|\hat{T}_{kl}^{(s')}|n\rangle$ , and when  $\langle n|\hat{T}_{ab}^{(u=s)}\sum_k|k\rangle$  is off-diagonal, and it can be paired to  $\langle m|\hat{T}_{ij}^{(s)}|n\rangle$ . Therefore it is required that  $k = m$ . Other three terms in the second step has similar results.

### B.3 Expansion details for $\chi_{ijkl}^{\text{super res}}$

$$\begin{aligned}
& \frac{2}{(N_0L)^3\hbar}\sum_{n^*l^*}\frac{e^{-\beta E_n^*}}{\mathcal{Z}^*}\langle l^*|\hat{T}_{ij,cc}|n^*\rangle\langle n^*|\hat{T}_{kl}|l^*\rangle\frac{\omega_{ln}^*}{(\omega+i\eta)^2-\omega_{ln}^{*2}} \\
= & \frac{2}{(N_0L)^3\hbar}\sum_{nl}\frac{e^{-\beta E_n}}{\mathcal{Z}}\langle l|\hat{T}_{ij,cc}|n\rangle\langle n|\hat{T}_{kl}|l\rangle\frac{\omega_{ln}}{(\omega+i\eta)^2-\omega_{ln}^2} \\
\text{term(7)} + & \frac{2}{(N_0L)^3\hbar}\sum_{nl}\frac{e^{-\beta E_n}(-\beta\delta E_n)}{\mathcal{Z}}\langle l|\hat{T}_{ij,cc}|n\rangle\langle n|\hat{T}_{kl}|l\rangle\frac{\omega_{ln}}{(\omega+i\eta)^2-\omega_{ln}^2} \\
\text{term(8)} + & \frac{2}{(N_0L)^3\hbar}\sum_{nl}\frac{e^{-\beta E_n}(-\delta\mathcal{Z})}{\mathcal{Z}}\langle l|\hat{T}_{ij,cc}|n\rangle\langle n|\hat{T}_{kl}|l\rangle\frac{\omega_{ln}}{(\omega+i\eta)^2-\omega_{ln}^2} \\
\text{term(9)} + & \frac{2}{(N_0L)^3\hbar}\sum_{nl}\frac{e^{-\beta E_n}}{\mathcal{Z}}\langle l|\hat{T}_{ij,cc}|n\rangle\langle n|\hat{T}_{kl}|l\rangle\frac{(\omega+i\eta)^2+\omega_{ln}^2}{[(\omega+i\eta)^2-\omega_{ln}^2]^2}\delta\omega_{ln} \\
\text{term(10)} + & \frac{2}{(N_0L)^3\hbar}\sum_{nl}\frac{e^{-\beta E_n}}{\mathcal{Z}}\left[\langle\delta|l\rangle\hat{T}_{ij,cc}|n\rangle\langle n|\hat{T}_{kl}|l\rangle+\langle l|\hat{T}_{ij,cc}(\delta|n)\rangle\langle n|\hat{T}_{kl}|l\rangle\right. \\
& \left.+\langle l|\hat{T}_{ij,cc}|n\rangle\langle\delta|n\rangle\hat{T}_{kl}|l\rangle+\langle l|\hat{T}_{ij,cc}|n\rangle\langle n|\hat{T}_{kl}(\delta|l)\rangle\right]\frac{\omega_{ln}}{(\omega+i\eta)^2-\omega_{ln}^2} \quad (\text{B.14})
\end{aligned}$$

where please note we use the simplified notation  $(E_l - E_n)/\hbar = \omega_l - \omega_n = \omega_{ln}$ . Thus the change of  $\omega_{ln}$ ,  $\delta\omega_{ln} = (\delta E_l - \delta E_n)/\hbar$ .

Expansion for term(7):

$$\begin{aligned}
& -\frac{2\beta}{\hbar(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \langle n|\hat{V}|n\rangle \langle l|\hat{T}_{ij,cc}|n\rangle \langle n|\hat{T}_{kl}|l\rangle \frac{\omega_{ln}}{(\omega+i\eta)^2-\omega_{ln}^2} \\
= & -\frac{2\beta}{\hbar(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik\cdot(x_s-x'_s)} \langle n|\hat{T}_{ab}^{(u)}\hat{T}_{cd}^{(u')}|n\rangle \langle l|\hat{T}_{ij}^{(s)}|n\rangle \langle n|\hat{T}_{kl}^{(s')}|l\rangle \frac{\omega_{ln}}{(\omega+i\eta)^2-\omega_{ln}^2} \\
= & -\frac{2\beta}{\hbar(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik\cdot(x_s-x'_s)} \\
& \langle n|\hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k|\hat{T}_{cd}^{(u')}|n\rangle \langle l|\hat{T}_{ij}^{(s)}|n\rangle \langle n|\hat{T}_{kl}^{(s')}|l\rangle \frac{\omega_{ln}}{(\omega+i\eta)^2-\omega_{ln}^2} \\
= & -\frac{4\beta}{\hbar(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-ik\cdot(x_s-x'_s)} \frac{\omega_{ln}}{(\omega+i\eta)^2-\omega_{ln}^2} \text{Tr} \left[ \hat{T}_{cd}^{(s)}|n\rangle \langle l|\hat{T}_{ij}^{(s)}|n\rangle \langle n|\hat{T}_{kl}^{(s')}|l\rangle \langle n|\hat{T}_{ab}^{(s')} \right] \\
= & -\frac{4\beta}{\hbar(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-ik\cdot(x_s-x'_s)} \frac{\omega_{ln}}{(\omega+i\eta)^2-\omega_{ln}^2} \\
& \langle n^{(s)}|\hat{T}_{ij}^{(s)}|n^{(s)}\rangle \langle n^{(s)}|\hat{T}_{ab}^{(s)}|n^{(s)}\rangle \langle n^{(s')}|\hat{T}_{cd}^{(s')}|n^{(s')}\rangle \langle n^{(s')}|\hat{T}_{kl}^{(s')}|n^{(s')}\rangle \langle n^{(s)}|l^{(s)}\rangle \langle n^{(s')}|l^{(s')}\rangle \langle n^{(r)}|l^{(r)}\rangle \\
= & 0
\end{aligned} \tag{B.15}$$

where in the above term's calculations we insert the identity  $\sum_k |k\rangle \langle k|$ . Since we have the coefficient  $\Lambda_{abcd}^{(uu')}$ ,  $u \neq u'$  so the matrix elements must be paired to stress tensors  $\hat{T}^{(s)}$  and  $\hat{T}^{(s')}$ . Since both of the matrix elements of  $\hat{T}^{(s)}$  and  $\hat{T}^{(s')}$  are off-diagonal, we need to pair the off-diagonal matrix elements for  $\hat{T}^{(u)}$  and  $\hat{T}^{(u')}$  with them. First we choose  $u = s'$  and  $u' = s$ . Since  $|n\rangle = \prod_s |n^{(s)}\rangle$  and the wave function at block position  $s$  has nothing to do with the operator which belongs to the block  $s'$ , we always have  $\hat{T}^{(s')}|n^{(s)}\rangle = |n^{(s)}\rangle \hat{T}^{(s')}$ . Thus we can rewrite  $\text{Tr} \left[ \hat{T}_{cd}^{(s)}|n\rangle \langle l|\hat{T}_{ij}^{(s)}|n\rangle \langle n|\hat{T}_{kl}^{(s')}|l\rangle \langle n|\hat{T}_{ab}^{(s')} \right]$  into

$$\text{Tr} \left[ \left( \hat{T}_{cd}^{(s)}|n^{(s)}\rangle \langle l^{(s)}|\hat{T}_{ij}^{(s)}|n^{(s)}\rangle \langle n^{(s)}|l^{(s)}\rangle \langle n^{(s)}| \right) \left( |n^{(s')}\rangle \langle l^{(s')}|n^{(s')}\rangle \langle n^{(s')}|\hat{T}_{kl}^{(s')}|l^{(s')}\rangle \langle n^{(s')}|\hat{T}_{ab}^{(s')} \right) \right]$$

we automatically get the constraints  $|n^{(s)}\rangle = |l^{(s)}\rangle$  and  $|n^{(s')}\rangle = |l^{(s')}\rangle$ . On the other hand we choose  $u = s$  and  $u' = s'$ , we have to rewrite  $\text{Tr} \left[ \hat{T}_{cd}^{(s')}|n\rangle \langle l|\hat{T}_{ij}^{(s)}|n\rangle \langle n|\hat{T}_{kl}^{(s')}|l\rangle \langle n|\hat{T}_{ab}^{(s)} \right]$  into

$$\text{Tr} \left[ \left( |n^{(s)}\rangle \langle l^{(s)}|\hat{T}_{ij}^{(s)}|n^{(s)}\rangle \langle n^{(s)}|l^{(s)}\rangle \langle n^{(s)}|\hat{T}_{ab}^{(s)} \right) \left( \hat{T}_{cd}^{(s')}|n^{(s')}\rangle \langle l^{(s')}|n^{(s')}\rangle \langle n^{(s')}|\hat{T}_{kl}^{(s')}|l^{(s')}\rangle \langle n^{(s')}| \right) \right]$$

we still automatically get the constraints  $|n^{(s)}\rangle = |l^{(s)}\rangle$  and  $|n^{(s')}\rangle = |l^{(s')}\rangle$ . With these constraints we finally reach the 0 result of the above term(7).

However, there is an additional qualitative argument which leads us to the same result for term(7) very quickly: suppose  $s \neq s'$  and  $l \neq n$  in the second step of Eq.(B.15). The operator for the  $s$ -th block  $\hat{T}^{(s)}$  changes state  $|l\rangle \rightarrow |n\rangle$  and the  $s'$ -th block operator  $\hat{T}^{(s')}$  changes state  $|n\rangle \rightarrow |l\rangle$ . However, since the  $s$ -th

block operator and the  $s'$ -th block operator only operate on wavefunctions which belong to the  $s$ -th block and  $s'$ -th block, it is impossible to change  $|l\rangle \rightarrow |n\rangle$  and  $|n\rangle \rightarrow |l\rangle$  simultaneously via two different block operators. The only possibility is  $|n\rangle = |l\rangle$ , which means the wave functions are not changed by both of  $\hat{T}^{(s)}$  and  $\hat{T}^{(s')}$ . This argument also leads to the same result of term(7), because the factor  $\omega_{ln} = \omega_l - \omega_n = 0$  makes term(7) to vanish.

Expansion for term(8):

$$\begin{aligned}
& -\frac{2}{\hbar(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}^2} \delta \mathcal{Z} \langle l | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | l \rangle \frac{\omega_{ln}}{(\omega + i\eta)^2 - \omega_{ln}^2} \\
&= \frac{2\beta}{\hbar(N_0L)^3} \sum_{lmn} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \langle m | V | m \rangle \langle l | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | l \rangle \frac{\omega_{ln}}{(\omega + i\eta)^2 - \omega_{ln}^2} \\
&= \frac{2\beta}{\hbar(N_0L)^3} \sum_{lmn} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
&\quad \frac{\omega_{ln}}{(\omega + i\eta)^2 - \omega_{ln}^2} \langle m | \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | m \rangle \langle l | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle \\
&= \frac{2\beta}{\hbar(N_0L)^3} \sum_{lmn} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
&\quad \frac{\omega_{ln}}{(\omega + i\eta)^2 - \omega_{ln}^2} \langle m | \hat{T}_{ab}^{(u)} \sum_k |k\rangle \langle k| \hat{T}_{cd}^{(u')} | m \rangle \langle l | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle \\
&= \frac{2\beta}{\hbar(N_0L)^3} \sum_{lmn} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \\
&\quad \frac{\omega_{ln}}{(\omega + i\eta)^2 - \omega_{ln}^2} \text{Tr} \left[ \hat{T}_{cd}^{(u')} | m \rangle \langle l | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle \langle m | \hat{T}_{ab}^{(u)} \right] \\
&= \frac{4\beta}{\hbar(N_0L)^3} \sum_{lmn} \frac{e^{-\beta(E_n+E_m)}}{\mathcal{Z}^2} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \frac{\omega_{ln}}{(\omega + i\eta)^2 - \omega_{ln}^2} \\
&\quad \langle m^{(s)} | \hat{T}_{cd}^{(s)} | m^{(s)} \rangle \langle l^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | n^{(s')} \rangle \langle m^{(s')} | \hat{T}_{ab}^{(s')} | m^{(s')} \rangle \langle n^{(s)} | l^{(s)} \rangle \langle n^{(s')} | l^{(s')} \rangle \langle n^{(r)} | l^{(r)} \rangle \\
&= 0
\end{aligned} \tag{B.16}$$

where again we insert the identity  $\sum_k |k\rangle \langle k|$  and use the same procedure and constraints as we mentioned in term(7) calculations.

Expansion for term(9):

$$\begin{aligned}
& \frac{2}{\hbar(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \langle l | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | l \rangle \frac{(\omega + i\eta)^2 + \omega_{ln}^2}{[(\omega + i\eta)^2 - \omega_{ln}^2]^2} \delta\omega_{ln} \\
&= \frac{2}{\hbar^2(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \langle l | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | l \rangle \frac{(\omega + i\eta)^2 + \omega_{ln}^2}{[(\omega + i\eta)^2 - \omega_{ln}^2]^2} \left( \langle l | \hat{V} | l \rangle - \langle n | \hat{V} | n \rangle \right) \\
&= \frac{2}{\hbar^2(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \frac{(\omega + i\eta)^2 + \omega_{ln}^2}{[(\omega + i\eta)^2 - \omega_{ln}^2]^2} \\
&\quad \langle l | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle \left( \langle l | \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | l \rangle - \langle n | \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | n \rangle \right) \\
&= \frac{2}{\hbar^2(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \frac{(\omega + i\eta)^2 + \omega_{ln}^2}{[(\omega + i\eta)^2 - \omega_{ln}^2]^2} \\
&\quad \left[ \text{Tr} \left( \hat{T}_{cd}^{(u')} | l \rangle \langle l | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle \langle l | \hat{T}_{ab}^{(u)} \right) - \text{Tr} \left( \hat{T}_{cd}^{(u')} | n \rangle \langle l | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle \langle n | \hat{T}_{ab}^{(u)} \right) \right] \\
&= \frac{1}{\hbar^2(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{abcd} \sum_{uu'} \sum_{ss'} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \frac{(\omega + i\eta)^2 + \omega_{ln}^2}{[(\omega + i\eta)^2 - \omega_{ln}^2]^2} \langle n^{(s)} | l^{(s)} \rangle \langle n^{(s')} | l^{(s')} \rangle \langle n^{(r)} | l^{(r)} \rangle \\
&\quad \left[ \langle n^{(s)} | \hat{T}_{ab}^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | n^{(s')} \rangle \right. \\
&\quad \left. - \langle n^{(s)} | \hat{T}_{ab}^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | n^{(s')} \rangle \right] \\
&= 0
\end{aligned} \tag{B.17}$$

where again we insert the identity  $\sum_k |k\rangle\langle k|$  and use the same procedure and constraints as we mentioned in term(7) calculations.

Expansion for term(10):

$$\begin{aligned}
& \frac{2}{\hbar(N_0L)^3} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \left[ (\delta\langle l |) \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} | l \rangle + \langle l | \hat{T}_{ij,cc} (\delta|n\rangle) \langle n | \hat{T}_{kl} | l \rangle \right. \\
&\quad \left. + \langle l | \hat{T}_{ij,cc} | n \rangle (\delta\langle n |) \hat{T}_{kl} | l \rangle + \langle l | \hat{T}_{ij,cc} | n \rangle \langle n | \hat{T}_{kl} (\delta|l\rangle) \right] \frac{\omega_{ln}}{(\omega + i\eta)^2 - \omega_{ln}^2} \\
&= \frac{2}{\hbar(N_0L)^3} \sum_{nl} \sum_{abcd} \sum_{uu'} \sum_{ss'} \frac{e^{-\beta E_n}}{\mathcal{Z}} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \frac{\omega_{ln}}{(\omega + i\eta)^2 - \omega_{ln}^2} \sum_m \\
&\quad \left\{ \frac{1}{E_l - E_m} \left( \langle l | \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | m \rangle \langle m | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle + \langle l | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | m \rangle \langle m | \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | l \rangle \right) \right. \\
&\quad \left. + \frac{1}{E_n - E_m} \left( \langle l | \hat{T}_{ij}^{(s)} | m \rangle \langle m | \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle + \langle l | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | m \rangle \langle m | \hat{T}_{kl}^{(s')} | l \rangle \right) \right\} \\
&= \frac{2}{\hbar(N_0L)^3} \sum_{nl} \sum_{abcd} \sum_{uu'} \sum_{ss'} \frac{e^{-\beta E_n}}{\mathcal{Z}} \Lambda_{abcd}^{(uu')} e^{-ik \cdot (x_s - x'_s)} \frac{\omega_{ln}}{(\omega + i\eta)^2 - \omega_{ln}^2} \sum_m \sum_k \\
&\quad \left\{ \frac{1}{E_l - E_m} \left( \langle l | \hat{T}_{ab}^{(u)} | k \rangle \langle k | \hat{T}_{cd}^{(u')} | m \rangle \langle m | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle + \langle l | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | m \rangle \langle m | \hat{T}_{ab}^{(u)} | k \rangle \langle k | \hat{T}_{cd}^{(u')} | l \rangle \right) \right. \\
&\quad \left. + \frac{1}{E_n - E_m} \left( \langle l | \hat{T}_{ij}^{(s)} | m \rangle \langle m | \hat{T}_{ab}^{(u)} | k \rangle \langle k | \hat{T}_{cd}^{(u')} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle + \langle l | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{ab}^{(u)} | k \rangle \langle k | \hat{T}_{cd}^{(u')} | m \rangle \langle m | \hat{T}_{kl}^{(s')} | l \rangle \right) \right\}
\end{aligned}$$

where in the third step of the above calculation we insert the identity  $\sum_k |k\rangle\langle k|$ . Because of the coefficient  $\Lambda_{abcd}^{(uu')}$ , we have  $u \neq u'$ . In the final result of the above Eq.(B.18) we get 4 summations. Let us discuss the first summation for example.

$$\frac{2}{\hbar N_0^3 V} \sum_{mnkl} \sum_{abcd} \sum_{uu'} \sum_{ss'} \frac{e^{-\beta E_n}}{\mathcal{Z}} \Lambda_{abcd}^{(uu')} \frac{\omega_{ln}}{(\omega + i\eta)^2 - \omega_{ln}^2} \frac{e^{-ik \cdot (x_s - x'_s)}}{E_l - E_m} \langle l | \hat{T}_{ab}^{(u)} | k \rangle \langle k | \hat{T}_{cd}^{(u')} | m \rangle \langle m | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle \quad (\text{B.19})$$

The pairing rule for the other three summations are the same. In the summation, Eq.(B.19), the matrix elements  $\langle l | \hat{T}_{ab}^{(u)} | k \rangle$ ,  $\langle k | \hat{T}_{cd}^{(u')} | m \rangle$  must be paired with  $\langle m | \hat{T}_{ij}^{(s)} | n \rangle$ ,  $\langle n | \hat{T}_{kl}^{(s')} | l \rangle$ . We get two candidates of pairing: first,  $\langle l | \hat{T}_{ab}^{(u)} | k \rangle$  is paired with  $\langle m | \hat{T}_{ij}^{(s)} | n \rangle$ , and  $\langle k | \hat{T}_{cd}^{(u')} | m \rangle$  is paired with  $\langle n | \hat{T}_{kl}^{(s')} | l \rangle$ ; second,  $\langle l | \hat{T}_{ab}^{(u)} | k \rangle$  is paired with  $\langle n | \hat{T}_{kl}^{(s')} | l \rangle$ , and  $\langle k | \hat{T}_{cd}^{(u')} | m \rangle$  is paired with  $\langle m | \hat{T}_{ij}^{(s)} | n \rangle$ .

In the first candidate, we have  $u = s, u' = s'$ . According to the factor  $\omega_{ln}/((i\eta)^2 - \omega_{ln}^2)$  which requires  $l \neq n$ , the matrix element  $\langle n | \hat{T}_{kl}^{(s')} | l \rangle$  must be off-diagonal. Therefore, the matrix element  $\langle k | \hat{T}_{cd}^{(u'=s')} | m \rangle$  which is paired to it must be off-diagonal as well. The wavefunctions of  $|k\rangle = \prod_{r=1}^{N_0^3} |k^{(r)}\rangle$  and  $|m\rangle = \prod_{r=1}^{N_0^3} |m^{(r)}\rangle$  are required to be  $|k^{(s')} \rangle = |l^{(s')} \rangle$ ,  $|m^{(s')} \rangle = |n^{(s')} \rangle$ , and  $\prod_{r \neq s'} |k^{(r)}\rangle = \prod_{r \neq s'} |m^{(r)}\rangle$ ,  $\prod_{r \neq s'} |n^{(r)}\rangle = \prod_{r \neq s'} |l^{(r)}\rangle$ . Therefore in the first candidate case, the first term in Eq.(B.19) is simplified as

$$\begin{aligned} & \frac{2}{\hbar N_0^3 V} \sum_{mnkl} \sum_{abcd} \sum_{ss'} \frac{e^{-\beta E_n}}{\mathcal{Z}} \Lambda_{abcd}^{(ss')} \frac{\omega_{ln}}{(\omega + i\eta)^2 - \omega_{ln}^2} \frac{e^{-ik \cdot (x_s - x'_s)}}{E_l - E_m} \langle l | \hat{T}_{ab}^{(s)} | k \rangle \langle m | \hat{T}_{ij}^{(s)} | n \rangle \langle k | \hat{T}_{cd}^{(s')} | m \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle \\ = & \frac{2}{\hbar N_0^3 V} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')} m^{(s)} n^{(s)} n^{(s')}} \frac{e^{-\beta(E_n^{(s)} + E_n^{(s')})}}{\mathcal{Z}^{(s)} \mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \frac{\omega_{ln}^{(s')}}{(\omega + i\eta)^2 - \omega_{ln}^{(s')2}} \\ & \frac{\langle n^{(s)} | \hat{T}_{ab}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle}{(E_n^{(s)} - E_m^{(s)}) + (E_l^{(s')} - E_n^{(s')})} \\ = & \frac{2}{\hbar N_0^3 V} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')} m^{(s)} n^{(s)} n^{(s')}} \frac{e^{-\beta(E_n^{(s)} + E_n^{(s')})}}{\mathcal{Z}^{(s)} \mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \frac{\omega_{ln}^{(s')}}{(\omega + i\eta)^2 - \omega_{ln}^{(s')2}} \\ & \frac{\langle n^{(s)} | \hat{T}_{cd}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{ab}^{(s')} | n^{(s')} \rangle}{(E_n^{(s)} - E_m^{(s)}) + (E_l^{(s')} - E_n^{(s')})} \end{aligned} \quad (\text{B.20})$$

where in the last step, we exchange the indices  $(ab)$  and  $(cd)$  in the stress tensors  $\hat{T}_{ab}^{(s)}$  and  $\hat{T}_{cd}^{(s')}$ . The exchange of indices is correct, because the coefficient  $\Lambda_{abcd}^{(ss')}$  have the symmetry property:  $\Lambda_{abcd}^{(ss')} = \Lambda_{cdab}^{(ss')}$ .

Next we consider the second candidate, with  $u = s', u' = s$ . Actually the second candidate equals to first candidate, because with the exchange of indices  $(ab)$ ,  $(cd)$  and  $(s)$ ,  $(s')$ , the coefficient  $\sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)}$  keeps invariant:  $\sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} = \sum_{ss'} \Lambda_{cdab}^{(s's)} e^{-ik \cdot (x'_s - x_s)}$ , and the stress tensor operators commute:  $[\hat{T}_{ab}^{(u)}, \hat{T}_{cd}^{(u')}]_{u \neq u'} = 0$ .

Repeat the same process for the other three summations in Eq.(B.20), we proceed our calculation of term(10) as follows,

$$\begin{aligned}
\text{term(10)} &= \frac{4}{\hbar(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')}m^{(s)}n^{(s)}n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)}\mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \frac{\omega_{ln}^{(s')}}{(\omega + i\eta)^2 - \omega_{ln}^{(s')2}} \\
&\quad \frac{\langle n^{(s)} | \hat{T}_{cd}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{ab}^{(s')} | n^{(s')} \rangle}{(E_n^{(s)} - E_m^{(s)}) + (E_l^{(s')} - E_n^{(s')})} \\
&+ \frac{4}{\hbar(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s)}m^{(s')}n^{(s)}n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)}\mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \frac{\omega_{ln}^{(s)}}{(\omega + i\eta)^2 - \omega_{ln}^{(s)2}} \\
&\quad \frac{\langle n^{(s)} | \hat{T}_{cd}^{(s)} | l^{(s)} \rangle \langle l^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | m^{(s')} \rangle \langle m^{(s')} | \hat{T}_{ab}^{(s')} | n^{(s')} \rangle}{(E_n^{(s')} - E_m^{(s')}) + (E_l^{(s)} - E_n^{(s)})} \\
&+ \frac{4}{\hbar(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')}m^{(s)}n^{(s)}n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)}\mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \frac{\omega_{ln}^{(s')}}{(\omega + i\eta)^2 - \omega_{ln}^{(s')2}} \\
&\quad \frac{\langle n^{(s)} | \hat{T}_{cd}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{ab}^{(s')} | n^{(s')} \rangle}{(E_n^{(s)} - E_m^{(s)}) - (E_l^{(s')} - E_n^{(s')})} \\
&+ \frac{4}{\hbar(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s)}m^{(s')}n^{(s)}n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)}\mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \frac{\omega_{ln}^{(s)}}{(\omega + i\eta)^2 - \omega_{ln}^{(s)2}} \\
&\quad \frac{\langle n^{(s)} | \hat{T}_{cd}^{(s)} | l^{(s)} \rangle \langle l^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | m^{(s')} \rangle \langle m^{(s')} | \hat{T}_{ab}^{(s')} | n^{(s')} \rangle}{(E_n^{(s')} - E_m^{(s')}) - (E_l^{(s)} - E_n^{(s)})} \tag{B.21}
\end{aligned}$$

There are 4 terms above. The 3rd and 4th terms are similar with the 1st and 2nd terms. Therefore let us focus on the first two terms: for the 1st term we exchange the indices  $l, m$  and  $s, s'$ , because  $\sum_{ss'} \Lambda_{ijkl} e^{ik \cdot (x_s - x'_s)} = \sum_{ss'} \Lambda_{ijkl} e^{-ik \cdot (x_s - x'_s)}$  and we assume  $\langle n | \hat{T} | m \rangle \langle m | \hat{T} | n \rangle$  is independent of the block



number. We find after the exchange of indices  $lm$  and  $ss'$  the 1st term is invariant:

$$\begin{aligned}
& \frac{4}{\hbar(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')} m^{(s)} n^{(s)} n^{(s')}} \sum_{n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)}\mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \frac{\omega_{ln}^{(s')}}{(\omega + i\eta)^2 - \omega_{ln}^{(s')2}} \\
& \frac{\langle n^{(s)} | \hat{T}_{cd}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{ab}^{(s')} | n^{(s')} \rangle}{(E_n^{(s)} - E_m^{(s)}) + (E_l^{(s')} - E_n^{(s')})} \\
= & \frac{4}{\hbar(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')} m^{(s)} n^{(s)} n^{(s')}} \sum_{n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)}\mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{ik \cdot (x_s - x'_s)} \frac{\omega_{mn}^{(s)}}{(\omega + i\eta)^2 - \omega_{mn}^{(s)2}} \\
& \frac{\langle n^{(s')} | \hat{T}_{cd}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{ij}^{(s')} | n^{(s')} \rangle \langle n^{(s)} | \hat{T}_{kl}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ab}^{(s)} | n^{(s)} \rangle}{(E_n^{(s')} - E_l^{(s')}) + (E_m^{(s)} - E_n^{(s)})} \\
= & \frac{4}{\hbar(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')} m^{(s)} n^{(s)} n^{(s')}} \sum_{n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)}\mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{ik \cdot (x_s - x'_s)} \frac{\omega_{nm}^{(s)}}{(\omega + i\eta)^2 - \omega_{nm}^{(s)2}} \\
& \frac{\langle n^{(s')} | \hat{T}_{cd}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{ij}^{(s')} | n^{(s')} \rangle \langle n^{(s)} | \hat{T}_{kl}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ab}^{(s)} | n^{(s)} \rangle}{(E_l^{(s')} - E_n^{(s')}) + (E_n^{(s)} - E_m^{(s)})} \tag{B.22}
\end{aligned}$$

Use the identity

$$\left( \frac{1}{\omega - x} - \frac{1}{\omega + y} \right) \frac{1}{x + y} = \frac{1}{\omega - x} \frac{1}{\omega + y} \tag{B.23}$$

and let us denote

$$\omega_{ln}^{(s')} = x \quad \omega_{nm}^{(s)} = y \tag{B.24}$$

and use the following identity

$$\begin{aligned}
\frac{\omega_{ln}^{(s')}}{(\omega + i\eta)^2 - \omega_{ln}^{(s')2}} &= \frac{1}{2} \left( \frac{1}{\omega + i\eta - \omega_{ln}^{(s')}} - \frac{1}{\omega + i\eta + \omega_{ln}^{(s')}} \right) \\
\frac{\omega_{nm}^{(s)}}{(\omega + i\eta)^2 - \omega_{nm}^{(s)2}} &= \frac{1}{2} \left( \frac{1}{\omega + i\eta - \omega_{nm}^{(s)}} - \frac{1}{\omega + i\eta + \omega_{nm}^{(s)}} \right) \tag{B.25}
\end{aligned}$$

we can use the above identities to derive

$$\left( \frac{1}{\omega + i\eta - \omega_{ln}^{(s')}} - \frac{1}{\omega + i\eta + \omega_{nm}^{(s)}} \right) \frac{1}{\omega_{ln}^{(s')} + \omega_{nm}^{(s)}} = \frac{1}{\omega + i\eta - \omega_{ln}^{(s')}} \frac{1}{\omega + i\eta + \omega_{nm}^{(s)}} \tag{B.26}$$

Finally the 1st term equals to

$$\begin{aligned}
& \frac{1}{\hbar^2(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')} m^{(s)} n^{(s)} n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)} \mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \\
& \langle n^{(s)} | \hat{T}_{cd}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{ab}^{(s')} | n^{(s')} \rangle \\
& \left[ \frac{1}{(\omega + i\eta - \omega_{ln}^{(s')})} \frac{1}{(\omega + i\eta + \omega_{nm}^{(s)})} + \frac{1}{(\omega + i\eta + \omega_{ln}^{(s')})} \frac{1}{(\omega + i\eta - \omega_{nm}^{(s)})} \right]
\end{aligned} \tag{B.27}$$

Similarly the 2nd term is

$$\begin{aligned}
& \frac{1}{\hbar^2(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')} m^{(s)} n^{(s)} n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)} \mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \\
& \langle n^{(s)} | \hat{T}_{cd}^{(s)} | l^{(s)} \rangle \langle l^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | m^{(s')} \rangle \langle m^{(s')} | \hat{T}_{ab}^{(s')} | n^{(s')} \rangle \\
& \left[ \frac{1}{(\omega + i\eta - \omega_{ln}^{(s)})} \frac{1}{(\omega + i\eta + \omega_{nm}^{(s')})} + \frac{1}{(\omega + i\eta + \omega_{ln}^{(s)})} \frac{1}{(\omega + i\eta - \omega_{nm}^{(s')})} \right]
\end{aligned} \tag{B.28}$$

3rd term,

$$\begin{aligned}
& -\frac{1}{\hbar^2(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')} m^{(s)} n^{(s)} n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)} \mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \\
& \langle n^{(s)} | \hat{T}_{cd}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{ab}^{(s')} | n^{(s')} \rangle \\
& \left[ \frac{1}{(\omega + i\eta + \omega_{ln}^{(s')})} \frac{1}{(\omega + i\eta + \omega_{nm}^{(s)})} + \frac{1}{(\omega + i\eta - \omega_{ln}^{(s')})} \frac{1}{(\omega + i\eta - \omega_{nm}^{(s)})} \right]
\end{aligned} \tag{B.29}$$

4th term

$$\begin{aligned}
& -\frac{1}{\hbar^2(N_0L)^3} \sum_{abcd} \sum_{ss'} \sum_{l^{(s')} m^{(s)} n^{(s)} n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)} \mathcal{Z}^{(s')}} \Lambda_{abcd}^{(ss')} e^{-ik \cdot (x_s - x'_s)} \\
& \langle n^{(s)} | \hat{T}_{cd}^{(s)} | l^{(s)} \rangle \langle l^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{kl}^{(s')} | m^{(s')} \rangle \langle m^{(s')} | \hat{T}_{ab}^{(s')} | n^{(s')} \rangle \\
& \left[ \frac{1}{(\omega + i\eta + \omega_{ln}^{(s)})} \frac{1}{(\omega + i\eta + \omega_{nm}^{(s')})} + \frac{1}{(\omega + i\eta - \omega_{ln}^{(s)})} \frac{1}{(\omega + i\eta - \omega_{nm}^{(s')})} \right]
\end{aligned} \tag{B.30}$$

Sum them up we obtain

$$\frac{V}{N_0^3} \sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{ik \cdot (x_s - x'_s)} \chi_{cdij}^{\text{res}}(\omega + i\eta) \chi_{klab}^{\text{res}}(\omega + i\eta) \tag{B.31}$$

The summation over space with respect to  $\Lambda_{abcd}^{(ss')}$  is:

$$-L_n^3 \sum_{ss'} \Lambda_{ijkl}^{(ss')} e^{-ik \cdot (x_s - x'_s)} = \frac{N_0^3}{\rho c_t^2} (\kappa_j \kappa_l \delta_{ik} - \kappa_i \kappa_j \kappa_k \kappa_l) \quad (\text{B.32})$$

where  $L_n$  is the unit block's dimension in the  $n$ -th step renormalization, and  $\vec{\kappa}$  is the unit vector of momentum  $\vec{k}$ .

## B.4 Derivations of the Effect of $\delta\hat{V}(t)$ 's contribution to Susceptibility Renormalization

Finally let's consider the higher order corrections to super block non-elastic susceptibility due to super block stress tensor correction in Eq.(3.16). There are two kinds of extra expansions in super block susceptibility, (1) the product between  $\hat{T}_{ij} = \sum_s e^{i\vec{k} \cdot x_s} \hat{T}_{ij}^{(s)}$  and  $\sum_{s \neq s'} \sum_{abcd} \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} e^{i\vec{k} \cdot (\vec{x}_s + \vec{x}'_s)/2} (\delta\Lambda_{abcd}^{(ss')}(\mathbf{e})) / \delta e_{ij}$ , and (2) the super block susceptibility expansion quadratic in the operator  $\sum_{s \neq s'} \sum_{abcd} \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} e^{i\vec{k} \cdot (\vec{x}_s + \vec{x}'_s)/2} (\delta\Lambda_{abcd}^{(ss')}(\mathbf{e})) / \delta e_{ij}$ . The susceptibility correction of the first kind is in odd orders of stress tensor matrix elements. Bare in mind that the stress tensors are a highly frustrated system, the expectation values of stress tensors are random quantities functional of spacial coordinates and it's quantum numbers  $(n, m)$  in  $\langle n | \hat{T}_{ij}^{(s)} | m \rangle$ . Those terms in odd orders of stress tensor matrix elements vanish after integrating over spacial coordinates, because it does not come out in pairs of stress tensors matrix element products. For the super block susceptibility expansion of the second kind, we calculate it's contribution to the first, second part of relaxation susceptibility, and the resonance susceptibility separately. The stress tensor for super block is by definition given by

$$\hat{T}_{ij} = \frac{\delta H(t)}{\delta e_{ij}(t)} = \sum_s e^{ik \cdot x_s} \hat{T}_{ij}^{(s)} + \sum_{ss'} e^{ik \cdot \frac{x_s + x'_s}{2}} \sum_{abcd} \frac{\delta \Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} \quad (\text{B.33})$$

where we choose the phonon strain field to be

$$e_{ij}^{(s)}(t) = e^{i(k \cdot x - \omega t)} e_{ij} \quad e_{ij}(t) = e^{-i\omega t} e_{ij} \quad (\text{B.34})$$

Which means it has a second contribution proportional to the quadratic in  $T_{ij}^{(s)}$  operators. Since the super block susceptibility by definition is given by

$$\begin{aligned} \chi_{ijkl}^{\text{super}}(\omega) &= \frac{1}{(N_0 L)^3} \frac{\beta}{1 - i\omega\tau^*} \left( \sum_{n^* m^*} \frac{e^{-\beta(E_n^* + E_m^*)}}{\mathcal{Z}^{*2}} \langle n^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle m^* | \hat{T}_{kl}^{\text{super}} | m^* \rangle \right. \\ &\quad \left. - \sum_{n^*} \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \langle n^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle n^* | \hat{T}_{kl}^{\text{super}} | n^* \rangle \right) \\ &+ \frac{1}{(N_0 L)^3} \frac{2}{\hbar} \sum_{n^* l^*} \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \langle l^* | \hat{T}_{ij,cc}^{\text{super}} | n^* \rangle \langle n^* | \hat{T}_{kl}^{\text{super}} | l^* \rangle \frac{\omega_l^* - \omega_n^*}{(\omega + i\eta)^2 - (\omega_l^* - \omega_n^*)^2} \quad (\text{B.35}) \end{aligned}$$

We need to take the term which is quadratic in  $\hat{T}_{ij}^{(s)}$  into account as well. Note that we are only interested in the 1st and 2nd order in susceptibility  $\chi$ , we only take quadratic and quatic order in  $\hat{T}_{ij}$  into account. The contribution from this quadratic operator term results in the change of susceptibility as follows,

$$\begin{aligned} &\frac{1}{(N_0 L)^3} \frac{\beta}{1 - i\omega\tau} \left( \sum_{nm} \frac{e^{-\beta(E_n + E_m)}}{\mathcal{Z}^2} \langle n | \sum_{ss'} e^{-i\left(k \cdot \frac{x_s + x'_s}{2}\right)} \sum_{abcd} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \right. \\ &\quad \left. \langle m | \sum_{uu'} e^{i\left(k \cdot \frac{x_u + x'_u}{2}\right)} \sum_{efgh} \frac{\delta\Lambda_{efgh}^{(uu')}}{\delta e_{kl}} \hat{T}_{ef}^{(u)} \hat{T}_{gh}^{(u')} | m \rangle \right) \\ &+ \frac{1}{(N_0 L)^3} \frac{\beta}{1 - i\omega\tau} \left( - \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \langle n | \sum_{ss'} e^{-i\left(k \cdot \frac{x_s + x'_s}{2}\right)} \sum_{abcd} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \right. \\ &\quad \left. \langle n | \sum_{uu'} e^{i\left(k \cdot \frac{x_u + x'_u}{2}\right)} \sum_{efgh} \frac{\delta\Lambda_{efgh}^{(uu')}}{\delta e_{kl}} \hat{T}_{ef}^{(u)} \hat{T}_{gh}^{(u')} | n \rangle \right) \\ &+ \frac{1}{(N_0 L)^3} \frac{2}{\hbar} \sum_{nl} \frac{e^{-\beta E_n}}{\mathcal{Z}} \frac{\omega_l - \omega_n}{(\omega + i\eta)^2 - (\omega_l - \omega_n)^2} \\ &\quad \langle l | \sum_{ss'} e^{-i\left(k \cdot \frac{x_s + x'_s}{2}\right)} \sum_{abcd} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \langle n | \sum_{uu'} e^{i\left(k \cdot \frac{x_u + x'_u}{2}\right)} \sum_{efgh} \frac{\delta\Lambda_{efgh}^{(uu')}}{\delta e_{kl}} \hat{T}_{ef}^{(u)} \hat{T}_{gh}^{(u')} | l \rangle \quad (\text{B.36}) \end{aligned}$$

Again, we calculate them one by one. The 1st term is

$$\begin{aligned}
& \frac{1}{(N_0 L)^3} \frac{\beta}{1 - i\omega\tau} \sum_{ss'uu'} \sum_{abcdefgh} \sum_{nm} \frac{e^{-\beta(E_n + E_m)}}{\mathcal{Z}^2} e^{-i\left(k \cdot \frac{x_s + x'_s}{2}\right)} e^{i\left(k \cdot \frac{x_u + x'_u}{2}\right)} \\
& \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(uu')}}{\delta e_{kl}} \langle n | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \langle m | \hat{T}_{ef}^{(u)} \hat{T}_{gh}^{(u')} | m \rangle \\
= & \frac{1}{(N_0 L)^3} \frac{\beta}{1 - i\omega\tau} \sum_{ss'} \sum_{abcdefgh} \sum_{nm} \frac{e^{-\beta(E_n + E_m)}}{\mathcal{Z}^2} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \langle n | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \langle m | \hat{T}_{ef}^{(s)} \hat{T}_{gh}^{(s')} | m \rangle \\
= & \frac{1}{(N_0 L)^3} \frac{\beta}{1 - i\omega\tau} \sum_{ss'} \sum_{abcdefgh} \sum_{nm} \frac{e^{-\beta(E_n + E_m)}}{\mathcal{Z}^2} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \\
& \langle n | \hat{T}_{ab}^{(s)} \sum_k |k\rangle \langle k| \hat{T}_{cd}^{(s')} | n \rangle \langle m | \hat{T}_{ef}^{(s)} \sum_{k'} |k'\rangle \langle k'| \hat{T}_{gh}^{(s')} | m \rangle \\
= & \frac{1}{(N_0 L)^3} \frac{\beta}{1 - i\omega\tau} \sum_{ss'} \sum_{abcdefgh} \sum_{nm} \frac{e^{-\beta(E_n + E_m)}}{\mathcal{Z}^2} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \\
& \sum_{kk'} \langle n^{(s)} | \hat{T}_{ab}^{(s)} | k^{(s)} \rangle \langle k^{(s)} | n^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ef}^{(s)} | k'^{(s)} \rangle \langle k'^{(s)} | m^{(s)} \rangle \\
& \langle n^{(s')} | k^{(s')} \rangle \langle k^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle \langle m^{(s')} | k'^{(s')} \rangle \langle k'^{(s')} | \hat{T}_{gh}^{(s')} | m^{(s')} \rangle \\
= & \frac{1}{(N_0 L)^3} \frac{\beta}{1 - i\omega\tau} \sum_{ss'} \sum_{abcdefgh} \sum_{n^{(s)} n^{(s')} m^{(s)} m^{(s')}} \frac{e^{-\beta(E_n^{(s)} + E_n^{(s')} + E_m^{(s)} + E_m^{(s')})}}{\mathcal{Z}^{(s)2} \mathcal{Z}^{(s')2}} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \\
& \langle n^{(s)} | \hat{T}_{ab}^{(s)} | n^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ef}^{(s)} | m^{(s)} \rangle \langle n^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle \langle m^{(s')} | \hat{T}_{gh}^{(s')} | m^{(s')} \rangle \\
= & \frac{L^3}{N_0^3} \frac{\beta^{-1}}{1 - i\omega\tau} \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \chi_{abef}^{\text{rel}(1)} \chi_{cdgh}^{\text{rel}(1)}
\end{aligned} \tag{B.37}$$

where in the above calculations we inserted the identities  $\sum_k |k\rangle \langle k|$  and  $\sum_{k'} |k'\rangle \langle k'|$ .

The 2nd term is

$$\begin{aligned}
& -\frac{1}{(N_0L)^3} \frac{\beta}{1-i\omega\tau} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ss'uu'} \sum_{abcdefgh} e^{-i\left(k \cdot \frac{x_s+x'_s}{2}\right)} e^{i\left(k \cdot \frac{x_u+x'_u}{2}\right)} \\
& \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(uu')}}{\delta e_{kl}} \langle n | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \langle n | \hat{T}_{ef}^{(u)} \hat{T}_{gh}^{(u')} | n \rangle \\
= & -\frac{1}{(N_0L)^3} \frac{\beta}{1-i\omega\tau} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \langle n | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \langle n | \hat{T}_{ef}^{(s)} \hat{T}_{gh}^{(s')} | n \rangle \\
= & -\frac{1}{(N_0L)^3} \frac{\beta}{1-i\omega\tau} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \\
& \langle n | \hat{T}_{ab}^{(s)} \sum_k |k\rangle \langle k| \hat{T}_{cd}^{(s')} | n \rangle \langle n | \hat{T}_{ef}^{(s)} \sum_{k'} |k'\rangle \langle k'| \hat{T}_{gh}^{(s')} | n \rangle \\
= & -\frac{1}{(N_0L)^3} \frac{\beta}{1-i\omega\tau} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \sum_{kk'} \\
& \langle n^{(s)} | \hat{T}_{ab}^{(s)} | k^{(s)} \rangle \langle k^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ef}^{(s)} | k'^{(s)} \rangle \langle k'^{(s)} | n^{(s)} \rangle \\
& \langle n^{(s')} | k^{(s')} \rangle \langle k^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle \langle n^{(s')} | k'^{(s')} \rangle \langle k'^{(s')} | \hat{T}_{gh}^{(s')} | n^{(s')} \rangle \\
= & -\frac{1}{(N_0L)^3} \frac{\beta}{1-i\omega\tau} \sum_{ss'} \sum_{n^{(s)}n^{(s')}} \frac{e^{-\beta(E_n^{(s)}+E_n^{(s')})}}{\mathcal{Z}^{(s)}\mathcal{Z}^{(s')}} \\
& \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \langle n^{(s)} | \hat{T}_{ab}^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ef}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle \langle n^{(s')} | \hat{T}_{gh}^{(s')} | n^{(s')} \rangle \\
= & -\frac{L^3}{N_0} \frac{\beta^{-1}}{1-i\omega\tau} \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \chi_{abef}^{\text{rel}(2)} \chi_{cdgh}^{\text{rel}(2)} \tag{B.38}
\end{aligned}$$

where in the above calculations we inserted the identities  $\sum_k |k\rangle \langle k|$  and  $\sum_{k'} |k'\rangle \langle k'|$ .

The 3rd term is

$$\begin{aligned}
& \frac{1}{(N_0 L)^3} \frac{1}{\hbar} \sum_{nl} (P_n - P_l) \sum_{ss'uu'} \sum_{abcdefgh} e^{-i\left(k \cdot \frac{x_s + x'_s}{2}\right)} e^{i\left(k \cdot \frac{x_u + x'_u}{2}\right)} \\
& \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(uu')}}{\delta e_{kl}} \frac{\langle l | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \langle n | \hat{T}_{ef}^{(u)} \hat{T}_{gh}^{(u')} | l \rangle}{\omega + i\eta + \omega_{nl}} \\
& = \frac{1}{(N_0 L)^3} \frac{1}{\hbar} \sum_{nl} (P_n - P_l) \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \frac{\langle l | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \langle n | \hat{T}_{ef}^{(s)} \hat{T}_{gh}^{(s')} | l \rangle}{\omega + i\eta + \omega_{nl}} \\
& = \frac{1}{(N_0 L)^3} \frac{1}{\hbar} \sum_{nl} (P_n - P_l) \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \frac{\langle l | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \langle n | \hat{T}_{ef}^{(s)} \hat{T}_{gh}^{(s')} | l \rangle}{\omega + i\eta + \omega_{nl}} \\
& = \frac{1}{(N_0 L)^3} \frac{1}{\hbar} \sum_{nl} (P_n - P_l) \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \\
& \quad \langle l | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \langle n | \hat{T}_{ef}^{(s)} \hat{T}_{gh}^{(s')} | l \rangle \left( \frac{1}{\omega + \omega_{nl}} - i\pi\delta(\omega + \omega_{nl}) \right) \\
& = \frac{1}{(N_0 L)^3} \frac{1}{\hbar} \sum_{nl} (P_n - P_l) \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \\
& \quad \langle l | \hat{T}_{ab}^{(s)} \sum_k |k\rangle \langle k | \hat{T}_{cd}^{(s')} | n \rangle \langle n | \hat{T}_{ef}^{(s)} \sum_{k'} |k'\rangle \langle k' | \hat{T}_{gh}^{(s')} | l \rangle \left( \frac{1}{\omega + \omega_{nl}} - i\pi\delta(\omega + \omega_{nl}) \right) \\
& = \frac{1}{(N_0 L)^3} \frac{1}{\hbar} \sum_{nl} (P_n - P_l) \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \left( \frac{1}{\omega + \omega_{nl}} - i\pi\delta(\omega + \omega_{nl}) \right) \\
& \quad \sum_{kk'} \langle l^{(s)} | \hat{T}_{ab}^{(s)} | k^{(s)} \rangle \langle k^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ef}^{(s)} | k'^{(s)} \rangle \langle k'^{(s)} | l^{(s)} \rangle \\
& \quad \langle l^{(s')} | k^{(s')} \rangle \langle k^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle \langle n^{(s')} | k'^{(s')} \rangle \langle k'^{(s')} | \hat{T}_{gh}^{(s')} | l^{(s')} \rangle \\
& = \frac{1}{(N_0 L)^3} \frac{1}{\hbar} \sum_{nl} (P_n - P_l) \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \\
& \quad \langle l^{(s)} | \hat{T}_{ab}^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ef}^{(s)} | l^{(s)} \rangle \langle l^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle \langle n^{(s')} | \hat{T}_{gh}^{(s')} | l^{(s')} \rangle \left( \frac{1}{\omega + \omega_{nl}} - i\pi\delta(\omega + \omega_{nl}) \right) \\
& = \frac{1}{(N_0 L)^3} \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \\
& \quad \sum_{n^{(s)} n^{(s')} l^{(s)} l^{(s')}} (P_n - P_l) \left( \frac{1}{\hbar\omega + \hbar\omega_{nl}^{(s)} + \hbar\omega_{nl}^{(s')}} - i\pi\delta(\hbar\omega + \hbar\omega_{nl}^{(s)} + \hbar\omega_{nl}^{(s')}) \right) \\
& \quad \int \langle n^{(s)} | \hat{T}_{ef}^{(s)} | l^{(s)} \rangle \langle l^{(s)} | \hat{T}_{ab}^{(s)} | n^{(s)} \rangle \delta(E_l^{(s)} - E_n^{(s)} - \hbar\omega_s) d(\hbar\omega_s) \\
& \quad \int \langle n^{(s')} | \hat{T}_{gh}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{cd}^{(s')} | n^{(s')} \rangle \delta(E_l^{(s')} - E_n^{(s')} - \hbar\omega'_s) d(\hbar\omega'_s) \\
& = \frac{L^3}{N_0^3 \pi^2} \sum_{ss'} \sum_{abcdefgh} \frac{\delta\Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta\Lambda_{efgh}^{(ss')}}{\delta e_{kl}} (1 - e^{-\beta\hbar(\omega_s + \omega'_s)}) \left( \frac{1}{\hbar\omega - \hbar\omega_s - \hbar\omega_{s'}} - i\pi\delta(\hbar\omega - \hbar\omega_s - \hbar\omega_{s'}) \right) \\
& \quad \int \frac{\text{Im} \chi_{abef}^{\text{res}}(\omega_s)}{1 - e^{-\beta\hbar\omega_s}} d(\hbar\omega_s) \int \frac{\text{Im} \chi_{cdgh}^{\text{res}}(\omega'_s)}{1 - e^{-\beta\hbar\omega'_s}} d(\hbar\omega'_s)
\end{aligned}$$

$$\begin{aligned}
&= -\frac{L^3}{N_0^3 \pi^2} \sum_{ss'} \sum_{abcdefgh} \frac{\delta \Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta \Lambda_{efgh}^{(ss')}}{\delta e_{kl}} \\
&\quad \int (1 - e^{-\beta \hbar (\omega_s + \omega'_s)}) \frac{\text{Im} \chi_{abef}^{\text{res}}(\omega_s) \text{Im} \chi_{cdgh}^{\text{res}}(\omega'_s)}{(1 - e^{-\beta \hbar \omega_s})(1 - e^{-\beta \hbar \omega'_s})(\hbar \omega_s + \hbar \omega'_s - \hbar \omega)} d(\hbar \omega_s) d(\hbar \omega'_s) \\
&\quad -i \frac{L^3}{N_0^3 \pi^2} \sum_{ss'} \sum_{abcdefgh} \frac{\delta \Lambda_{abcd}^{(ss')}}{\delta e_{ij}} \frac{\delta \Lambda_{efgh}^{(ss')}}{\delta e_{kl}} (1 - e^{-\beta \hbar \omega}) \left( \pi \int \frac{\text{Im} \chi_{abef}^{\text{res}}(\omega_s) \text{Im} \chi_{cdgh}^{\text{res}}(\omega - \omega_s)}{(1 - e^{-\beta \hbar \omega_s})(1 - e^{-\beta \hbar \omega'_s})} d(\hbar \omega_s) \right)
\end{aligned} \tag{B.39}$$

The super block susceptibility extra expansion terms Eq.(B.37, B.38, B.39) are the terms in the third and fourth lines of susceptibility renormalization equation.



# Appendix C

## Derivation Details of Resonance Phonon Energy Absorption in Generic Coupled Block Model

### C.1 Resonant Phonon Energy Absorption of Single Block Glass

In this chapter we want to give a detailed calculation on all of the resonant phonon energy absorption terms which appeared in chapter 6: Eq.(6.15, 6.20, 6.21, 6.22). First of all, in this section we calculate single block phonon energy absorption. Consider a single block of glass with the size  $L$  and non-elastic Hamiltonian  $\hat{H}$ . As we have mentioned earlier in chapter 6, Eq.(6.6), we can expand non-elastic Hamiltonian in orders of phonon long-wavelength strain field  $e_{ij}(\vec{x})$ :

$$\hat{H} = \hat{H}_0 + \int d^3x \sum_{ij} e_{ij}(\vec{x}) \hat{T}_{ij}(\vec{x}) + \mathcal{O}(e_{ij}^2) \quad (\text{C.1})$$

We define the eigenvalues and eigenstates for unperturbed non-elastic Hamiltonian  $\hat{H}_0$  to be  $E_n$  and  $|n\rangle$ . The genetic multiple-level-system  $\hat{H}_0$  can resonantly absorb phonon energy when a certain pair of energy levels  $E_n - E_m$  matches  $\hbar\omega$ . At the same time, such a pair of eigenstates  $|n\rangle, |m\rangle$  spontaneously emit phonon energy. Taking both of the emission/absorption processes into account, the net resonant phonon energy absorption is given by considering the entire set of eigenstates of  $\hat{H}_0$ , and by using Fermi golden rule:

$$E^{\text{single}}(t) = \frac{2\pi\omega t}{\hbar} \sum_{nm} \frac{e^{-\beta E_n}}{\mathcal{Z}} |\langle m | \sum_{ij} e_{ij} \hat{T}_{ij} | n \rangle|^2 \delta(E_n - E_m - \hbar\omega) \quad (\text{C.2})$$

where  $\mathcal{Z} = \sum_m e^{-\beta E_m}$  is the partition function for unperturbed non-elastic Hamiltonian  $\hat{H}_0$ . In the rest of this chapter we use the simplification  $\hbar = 1$ . To calculate Eq.(C.2) we need to make use of imaginary non-elastic resonance susceptibility, with the definition given in Eq.(6.7, 6.10),

$$\begin{aligned} \text{Im} \chi_{ijkl}^{\text{res}}(T, \omega) &= \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \text{Im} \chi_{ijkl}^{(m)}(\omega) \\ \text{Im} \chi_{ijkl}^{(m)}(\omega) &= \frac{\pi}{L^3} \int d^3x d^3x' \sum_n \langle m | \hat{T}_{ij}(\vec{x}) | n \rangle \langle n | \hat{T}_{kl}(\vec{x}') | m \rangle \\ &\quad [-\delta(E_n - E_m - \omega) + \delta(E_n - E_m + \omega)] \end{aligned} \quad (\text{C.3})$$

Please note, that the imaginary part of resonance non-elastic susceptibility is negative-definite. This definition is self-consistent with the definition of non-elastic susceptibility in chapters 4 and 5. Also, it is convenient to rewrite the imaginary resonance susceptibility Eq.(C.3) into reduced imaginary susceptibility  $\text{Im } \tilde{\chi}_{ijkl}^{\text{res}}$  as follows for future use:

$$\begin{aligned}
\text{Im } \chi_{ijkl}^{\text{res}}(T, \omega) &= (1 - e^{-\beta\hbar\omega}) \text{Im } \tilde{\chi}_{ijkl}^{\text{res}}(T, \omega) \\
\text{Im } \tilde{\chi}_{ijkl}^{\text{res}}(T, \omega) &= \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \text{Im } \tilde{\chi}_{ijkl}^{(m)}(\omega) \\
\text{Im } \tilde{\chi}_{ijkl}^{(m)}(\omega) &= -\frac{\pi}{L^3} \int d^3x d^3x' \sum_n \langle m | \hat{T}_{ij}(\vec{x}) | n \rangle \langle n | \hat{T}_{kl}(\vec{x}') | m \rangle \delta(E_n - E_m - \omega)
\end{aligned} \tag{C.4}$$

For an arbitrary isotropic system the reduced non-elastic susceptibility must satisfy the genetic form

$$\text{Im } \tilde{\chi}_{ijkl}^{\text{res}}(T, \omega) = (\text{Im } \tilde{\chi}_l^{\text{res}}(T, \omega) - 2 \text{Im } \tilde{\chi}_t^{\text{res}}(T, \omega)) \delta_{ij} \delta_{kl} + \text{Im } \tilde{\chi}_t^{\text{res}}(T, \omega) (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) \tag{C.5}$$

where please note we use  $\text{Im } \tilde{\chi}_{l,t}^{\text{res}}(T, \omega)$  to stand for imaginary part of reduced non-elastic longitudinal transverse susceptibility  $\text{Im } \tilde{\chi}_{l,t}^{\text{res non}}(T, \omega)$ . By definition they are negative quantities. The real part of reduced non-elastic susceptibility  $\text{Re } \tilde{\chi}_{ijkl}^{\text{res}}(T, \omega)$  can be obtained by Kramers-Kronig relation from the imaginary part of it. With the above definitions, we can directly calculate Eq.(C.2) to obtain resonant phonon energy absorption per unit time in single block glass

$$\dot{E}_{l,t}^{\text{single}} = -2L^3 A^2 k^2 \omega (1 - e^{-\beta\hbar\omega}) \text{Im } \tilde{\chi}_{l,t}^{\text{res}}(T, \omega) \tag{C.6}$$

This term appears in Eq.(6.15) in chapter 6. Again, the reduced version of the imaginary part of resonance susceptibility is negative definite. So the ‘‘energy absorption’’ for a single block glass is always positive. This result is intuitively correct.

## C.2 Resonant Phonon Energy Absorption of Super Block Glass

Now let’s combine  $N_0^3 L \times L \times L$  single blocks to form a  $N_0 L \times N_0 L \times N_0 L$  super block, and turn on virtual phonon exchange interactions between these single blocks:  $\hat{V} = \sum_{s \neq s'} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}$ . From chapter 6 the super block non-elastic Hamiltonian is given by the ‘‘super block unperturbed Hamiltonian  $\hat{H}_0^{\text{super}}$ ’’, which is static, and the ‘‘super block time-dependent perturbation  $\hat{H}'(t)$ ’’, which is the summation of (1) stress-strain coupling, (2) the modification of  $\Lambda_{ijkl}^{(ss')}$  due to external real phonon strain field, and (3) the modification of

stress tensor  $\hat{T}_{ij}$  via external phonon strain:

$$\begin{aligned}
\hat{H}^{\text{super}}(\mathbf{e}) &= \hat{H}_0^{\text{super}} + \hat{H}'(t) \\
\hat{H}_0^{\text{super}} &= \sum_s^{N_0^3} \hat{H}_0^{(s)} + \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \\
\hat{H}'(t) &= \sum_s^{N_0^3} \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)} + \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \left( \Delta \Lambda_{ijkl}^{(ss')} (t) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} + 2\Lambda_{ijkl}^{(ss')} \Delta \hat{T}_{ij}^{(s)}(t) \hat{T}_{kl}^{(s')} \right) \quad (\text{C.7})
\end{aligned}$$

for details of getting this Hamiltonian, please see chapter 6. We apologize for so many unnecessary definitions, but if not, the following calculations and equations will be super lengthy. We require a further definition for future simplicity, that the “change of virtual phonon exchange interaction  $\sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \left( \Delta \Lambda_{ijkl}^{(ss')} (t) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} + 2\Lambda_{ijkl}^{(ss')} \Delta \hat{T}_{ij}^{(s)}(t) \hat{T}_{kl}^{(s')} \right)$ ” is denoted as  $\delta \hat{V}(t)$ :

$$\begin{aligned}
\delta \hat{V}(t) &= \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \left( \Delta \Lambda_{ijkl}^{(ss')} (t) \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} + 2\Lambda_{ijkl}^{(ss')} \Delta \hat{T}_{ij}^{(s)}(t) \hat{T}_{kl}^{(s')} \right) \\
\Rightarrow \hat{H}'(t) &= \sum_s^{N_0^3} \sum_{ij} e_{ij}^{(s)}(t) \hat{T}_{ij}^{(s)} + \delta \hat{V}(t) \quad (\text{C.8})
\end{aligned}$$

Our main purpose here is to calculate resonant phonon energy absorption from the “super block time-dependent perturbation  $\hat{H}'(t)$ ”. If we define  $E_n$  and  $|n\rangle$  to be the eigenvalues and eigenstates of single block unperturbed Hamiltonian  $\sum_s^{N_0^3} \hat{H}_0^{(s)}$ , and  $E_n^*$  and  $|n^*\rangle$  to be the eigenstates and eigenvalues for super block unperturbed Hamiltonian,  $\sum_s^{N_0^3} \hat{H}_0^{(s)} + \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')}$ , the relations between  $E_n$  and  $E_n^*$ , and  $|n\rangle$  and  $|n^*\rangle$  are given by

$$|n^*\rangle = |n\rangle + \sum_{l \neq n} \frac{\langle l | \hat{V} | n \rangle}{E_n - E_l} |l\rangle + \dots \quad E_n^* = E_n + \langle n | \hat{V} | n \rangle + \sum_{l \neq n} \frac{|\langle l | \hat{V} | n \rangle|^2}{E_n - E_l} + \dots \quad (\text{C.9})$$

where we assume virtual phonon exchange interaction  $\hat{V}$  is relatively weak compared to single block unperturbed Hamiltonian  $\sum_s \hat{H}_0^{(s)}$ , so  $\hat{V}$  can be treated as a static perturbation. The formal way to calculate resonant phonon energy absorption via Hamiltonian  $\hat{H}_0^{\text{super}} + \hat{H}'(t)$  is to use interaction picture. For arbitrary operator  $\hat{A}$  and wave function  $|n^*\rangle$ , their interaction picture version are given by

$$\begin{aligned}
\hat{A}_I(t) &= e^{i\hat{H}_0^{\text{super}}t/\hbar} \hat{A} e^{-i\hat{H}_0^{\text{super}}t/\hbar} \\
|n_I^*, t\rangle &= e^{-\frac{i}{\hbar} \int_{-\infty}^t \hat{H}'(t') dt'} |n^*, t\rangle
\end{aligned} \tag{C.10}$$

where please be careful that in the definition of wave function interaction picture  $|n_I^*, t\rangle$ , the perturbation  $\hat{H}'_I(t')$  is also the interaction picture version of  $\hat{H}'(t)$ .

After all of the above preparations, finally the formal form of resonant phonon energy absorption per unit time is given by

$$\dot{E}_{l,t}^{\text{super}}(L) = \frac{\partial}{\partial t} \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \left( \langle n_I^*, t | \hat{H}_{0I}(t) + \hat{V}_I(t) | n_I^*, t \rangle - \langle n^* | \hat{H}_0 + \hat{V} | n^* \rangle \right) \tag{C.11}$$

where  $\mathcal{Z}^* = \sum_n e^{-\beta E_n^*}$  is the partition function for super block unperturbed Hamiltonian  $\hat{H}_0^{\text{super}}$ . Let us expand the first term in Eq.(C.11),  $\sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \langle n_I^*, t | \hat{H}_{0I}(t) + \hat{V}_I(t) | n_I^*, t \rangle$  up to the second order in phonon strain field.

$$\begin{aligned}
& \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \langle n_I^*, t | \hat{H}_{0I}(t) + \hat{V}_I(t) | n_I^*, t \rangle \\
&= \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \langle n^* | e^{-\frac{i}{\hbar} \int^t (\sum_{ij} \sum_s e_{ij,I}^{(s)}(t') \hat{T}_{ij,I}^{(s)}(t') + \delta \hat{V}_I(t')) dt'} \\
& \quad \left( \sum_s \hat{H}_{0,I}^{(s)}(t) + \hat{V}_I(t) \right) e^{\frac{i}{\hbar} \int^t (\sum_{ij} \sum_s e_{ij,I}^{(s)}(t'') \hat{T}_{ij,I}^{(s)}(t'') + \delta \hat{V}_I(t'')) dt''} | n^* \rangle \\
&= \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} E_n^* + \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \int^t dt' dt'' \sum_{l^*} e^{-i(E_l^* - E_n^*)(t' - t'')/\hbar} (E_l^* - E_n^*) \\
& \quad \langle n^* | \sum_{ij} \sum_s e_{ij}^{(s)}(t') \hat{T}_{ij}^{(s)} + \delta \hat{V}(t') | l^* \rangle \langle l^* | \sum_{ij} \sum_{s'} e_{ij}^{(s')}(t'') \hat{T}_{ij}^{(s')} + \delta \hat{V}(t'') | n^* \rangle \\
&= \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} E_n^* \\
\text{term[1]} &+ \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \int^t dt' dt'' \sum_{l^*} e^{-i(E_l^* - E_n^*)(t' - t'')/\hbar} (E_l^* - E_n^*) \\
& \quad \langle n^* | \sum_{ij} \sum_s e_{ij}^{(s)}(t') \hat{T}_{ij}^{(s)} | l^* \rangle \langle l^* | \sum_{ij} \sum_{s'} e_{ij}^{(s')}(t'') \hat{T}_{ij}^{(s')} | n^* \rangle \\
\text{term[2]} &+ \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \int^t dt' dt'' \sum_{l^*} e^{-i(E_l^* - E_n^*)(t' - t'')/\hbar} (E_l^* - E_n^*) \langle n^* | \delta \hat{V}(t') | l^* \rangle \langle l^* | \delta \hat{V}(t'') | n^* \rangle
\end{aligned} \tag{C.12}$$

where in the last step of the above calculation, we did not take the cross-over term between  $\delta\hat{V}$  and  $\sum_{ij} e_{ij}\hat{T}_{ij}$  into account, because we assume that the expectation value of three stress tensor product,  $\langle n|\hat{T}_{ij}\hat{T}_{kl}\hat{T}_{mn}|n\rangle$  vanishes if we average over the randomness of glass. The phonon resonance energy absorption, Eq.(C.11) is therefore simplified as  $\dot{E}_{l,t}^{\text{super}}(L) = \partial_t (\text{term}[1] + \text{term}[2])$ . Our main purpose of this chapter is to calculate term[1] and term[2] then.

### C.2.1 Calculation Details of Term[1]: Eq.(6.15, 6.22) in Chapter 6

We expand term[1] up to the second order of phonon strain field. Please note that we are only interested in the terms up to the second order of non-elastic susceptibility.

$$\begin{aligned}
\text{term}[1] &= \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \int^t dt' dt'' \sum_{l^*} e^{-i(E_l^* - E_n^*)(t' - t'')/\hbar} (E_l^* - E_n^*) \\
&\quad \langle n^* | \sum_{ij} \sum_s e_{ij}^{(s)}(t') \hat{T}_{ij}^{(s)} | l^* \rangle \langle l^* | \sum_{ij} \sum_{s'} e_{ij}^{(s')}(t'') \hat{T}_{ij}^{(s')} | n^* \rangle \\
&= \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} \sum_{l^*} (E_l^* - E_n^*) \langle n^* | \hat{T}_{ij}^{(s)} | l^* \rangle \langle l^* | \hat{T}_{kl}^{(s')} | n^* \rangle \\
&\quad \int^t dt' dt'' e^{-i(\omega_l^* - \omega_n^*)(t' - t'')/\hbar} \left[ e^{i(\omega l' - kx_s) - i(\omega t'' - kx'_s)} + e^{-i(\omega t' - kx_s) + i(\omega t'' - kx'_s)} \right] \\
&= \frac{2\pi\omega t}{\hbar} \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} \sum_{l^*} \langle n^* | \hat{T}_{ij}^{(s)} | l^* \rangle \langle l^* | \hat{T}_{kl}^{(s')} | n^* \rangle e^{-ik \cdot (x_s - x'_s)} \\
&\quad \left[ \frac{1}{\pi} \text{Im} \left( \frac{1}{\omega - \omega_l^* + \omega_n^* - i\eta} \right) - \frac{1}{\pi} \text{Im} \left( \frac{1}{\omega + \omega_l^* - \omega_n^* - i\eta} \right) \right] \tag{C.13}
\end{aligned}$$

where we define  $\omega_n = E_n/\hbar$  and  $\omega_n^* = E_n^*/\hbar$ . Next we apply the relations Eq.(C.9). Please note that the expectation value of virtual phonon exchange interaction  $\langle n|\hat{V}|n\rangle$  is always zero when  $|n\rangle = \prod_s |n^{(s)}\rangle$  stands for the eigenstates for single block Hamiltonians  $\sum_s \hat{H}_0^{(s)}$ . Therefore, in Eq.(C.9) the first order correction to  $E_n^*$  ( $\omega_n^*$ ) always vanishes. We need to expand  $E_n^*$  ( $\omega_n^*$ ) to the second order in  $\hat{V}$  to calculate the imaginary part of  $(\omega - \omega_l^* + \omega_n^* - i\eta)^{-1}$  to calculate Eq.(C.13). However, if we try to expand  $(\omega - \omega_l^* + \omega_n^* - i\eta)^{-1}$  in orders of  $\hat{V}$ , the lowest order is the second order expansion. Together with the term  $\langle n^* | \hat{T}_{ij}^{(s)} | l^* \rangle \langle l^* | \hat{T}_{kl}^{(s')} | n^* \rangle$  in the last step of Eq.(C.13), we will get a term written in the third order of non-elastic susceptibility. Because we are only interested in the terms up to the second order of non-elastic susceptibility, the order of  $(\omega - \omega_l^* + \omega_n^* - i\eta)^{-1}$  expansion is too high. Hence in the following calculation we use the approximation

$$\begin{aligned}
\frac{1}{\pi} \text{Im} \left( \frac{1}{\omega - \omega_l^* + \omega_n^* - i\eta} \right) &\approx \frac{1}{\pi} \text{Im} \left( \frac{1}{\omega - \omega_l + \omega_n - i\eta} \right) \\
\frac{1}{\pi} \text{Im} \left( \frac{1}{\omega + \omega_l^* - \omega_n^* - i\eta} \right) &\approx \frac{1}{\pi} \text{Im} \left( \frac{1}{\omega + \omega_l - \omega_n - i\eta} \right) \tag{C.14}
\end{aligned}$$

We further apply the approximation that  $\frac{1}{\pi} \text{Im} \left( \frac{1}{\omega - \omega_l + \omega_n - i\eta} \right) \approx \delta(\omega - \omega_l + \omega_n)$  and  $\frac{1}{\pi} \text{Im} \left( \frac{1}{\omega + \omega_l - \omega_n - i\eta} \right) \approx \delta(\omega + \omega_l - \omega_n)$ . On the hand, the  $n^*$ -th level probability  $e^{-\beta E_n^*} / \mathcal{Z}^*$  can be expanded in orders of  $\hat{V}$  as well. However, since both of  $e^{-\beta E_n^*}$  and  $\mathcal{Z}^*$  expansions come from the higher order corrections of  $E_n^*$ , the lowest order expansion for  $e^{-\beta E_n^*} / \mathcal{Z}^*$  is in the second order of  $\hat{V}$ . Combining with  $\langle n^* | \hat{T}_{ij}^{(s)} | l^* \rangle \langle l^* | \hat{T}_{kl}^{(s')} | n^* \rangle$  we still get a term which is in the third order of non-elastic susceptibility. Therefore we use the approximation that  $e^{-\beta E_n^*} / \mathcal{Z}^* \approx e^{-\beta E_n} / \mathcal{Z}$ . Therefore Eq.(C.13) can be further simplified as

$$\frac{2\pi\omega t}{\hbar} (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} \sum_{l^*} \langle n^* | \hat{T}_{ij}^{(s)} | l^* \rangle \langle l^* | \hat{T}_{kl}^{(s')} | n^* \rangle e^{-ik \cdot (x_s - x_{s'})} \delta(\omega - \omega_l + \omega_n) \quad (\text{C.15})$$

then we continue the calculation of Eq.(C.15):

$$\begin{aligned}
& \frac{2\pi\omega t}{\hbar} (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} \sum_{l^*} \langle n^* | \hat{T}_{ij}^{(s)} | l^* \rangle \langle l^* | \hat{T}_{kl}^{(s')} | n^* \rangle \delta(\omega - \omega_l + \omega_n) e^{-ik \cdot (r_s - r'_s)} \\
= & \frac{2\pi\omega t}{\hbar} (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} \sum_l \\
& \left( \langle n | + \sum_m \frac{\langle n | \hat{V} | m \rangle}{E_n - E_m} \langle m | \right) | \hat{T}_{ij}^{(s)} \left( | l \rangle + \sum_p \frac{\langle p | \hat{V} | l \rangle}{E_l - E_p} | p \rangle \right) \\
& \left( \langle l | + \sum_N \frac{\langle l | \hat{V} | N \rangle}{E_l - E_N} \langle N | \right) | \hat{T}_{kl}^{(s')} \left( | n \rangle + \sum_M \frac{\langle M | \hat{V} | n \rangle}{E_n - E_M} | M \rangle \right) \delta(\omega - \omega_l + \omega_n) e^{-ik \cdot (r_s - r'_s)} \\
\text{term(1)} = & 2\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_l \langle n | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | n \rangle \delta(\hbar\omega - E_l + E_n) \\
\text{term(2)} + & 2\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_{lm} \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 \delta(\hbar\omega - E_l + E_n) \\
\text{term(3)} + & 2\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_{lm} \langle n | \hat{T}_{ij}^{(s)} | m \rangle \frac{\langle m | \hat{V} | l \rangle}{E_l - E_m} \langle l | \hat{T}_{kl}^{(s')} | n \rangle \delta(\hbar\omega - E_l + E_n) \\
\text{term(4)} + & 2\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_{lm} \langle n | \hat{T}_{ij}^{(s)} | l \rangle \frac{\langle l | \hat{V} | m \rangle}{E_l - E_m} \langle m | \hat{T}_{kl}^{(s')} | n \rangle \delta(\hbar\omega - E_l + E_n) \\
\text{term(5)} + & 2\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_{lm} \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} | m \rangle \delta(\hbar\omega - E_l + E_n) \tag{C.16}
\end{aligned}$$

There are 5 terms need to be calculated. Term(1) is actually single block glass phonon energy absorption.

We have already calculated it before:

$$\begin{aligned}
& 2\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \sum_l \langle n | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | n \rangle \delta(\hbar\omega - E_l + E_n) \\
= & -2N_0^3 L^3 A^2 k^2 \omega (1 - e^{-\beta\hbar\omega}) \text{Im} \tilde{\chi}_{l,t}^{\text{res}}(T, \omega) \tag{C.17}
\end{aligned}$$

Again, Eq.(C.17) is positive-definite. This is Eq.(6.15) in chapter 6. Term(4) equals to term(2), and term(5) equals to term(3). Therefore we are only required to calculate term(2) and term(3). Term(2):

$$\begin{aligned}
& 2\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \sum_{lm} \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 \delta(\hbar\omega - E_l + E_n) \\
= & 2\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijklabcd} \sum_{ss'uu'} \Lambda_{abcd}^{(uu')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_{lm} \frac{\delta(\hbar\omega - E_l + E_n)}{E_n - E_m} \langle n | \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | m \rangle \langle m | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | n \rangle \\
= & 4\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_{lmp} \frac{\delta(\hbar\omega - E_l + E_n)}{E_n - E_m} \langle n | \hat{T}_{ab}^{(s')} | p \rangle \langle p | \hat{T}_{cd}^{(s)} | m \rangle \langle m | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | n \rangle \\
= & 4\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_{lm} \frac{\delta(\hbar\omega - E_l + E_n)}{E_n - E_m} \text{Tr} \left( \hat{T}_{cd}^{(s)} | m \rangle \langle m | \hat{T}_{ij}^{(s)} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | n \rangle \langle n | \hat{T}_{ab}^{(s')} \right) \\
= & 4\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_{l^{(s)} l^{(s')} l^{(r)} m^{(s)} m^{(s')} m^{(r)}} \frac{\delta(\hbar\omega - E_l^{(s)} - E_l^{(s')} - \sum E_l^{(r)} + E_n^{(s)} + E_n^{(s')} + \sum E_n^{(r)})}{E_n^{(s)} + E_n^{(s')} + \sum E_n^{(r)} - E_m^{(s)} - E_m^{(s')} - \sum E_m^{(r)}} \\
& \text{Tr} \prod_{r \neq s, s'} \left( |m^{(r)}\rangle \langle m^{(r)}| l^{(r)} \rangle \langle l^{(r)}| n^{(r)} \rangle \langle n^{(r)}| \right) \\
& \text{Tr} \left( \hat{T}_{cd}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | l^{(s)} \rangle \langle l^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \right) \text{Tr} \left( |m^{(s')} \rangle \langle m^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{kl}^{(s')} | n^{(s')} \rangle \langle n^{(s')} | \hat{T}_{ab}^{(s')} \right) \\
= & 4\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \sum_{l^{(s')} m^{(s)}} \frac{\delta(\hbar\omega - E_l^{(s')} + E_n^{(s')})}{E_n^{(s)} - E_m^{(s)} - \hbar\omega} \\
& \langle n^{(s)} | \hat{T}_{cd}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s')} | \hat{T}_{ab}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{kl}^{(s')} | n^{(s')} \rangle \\
= & 4 \frac{L_0^3}{\pi} \pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \sum_{n^{(s')}} \frac{e^{-\beta E_n^{(s')}}}{\mathcal{Z}^{(s')}} \text{Im} \tilde{\chi}_{abkl}^{(n_s')}(\omega) \\
& \sum_{n^{(s)}} \frac{e^{-\beta E_n^{(s)}}}{\mathcal{Z}^{(s)}} \sum_{m^{(s)}} \frac{1}{E_m^{(s)} - E_n^{(s)} + \hbar\omega} \langle n^{(s)} | \hat{T}_{cd}^{(s)} | m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \\
= & -4 \frac{L_0^6}{\pi} \omega t (1 - e^{-\beta\hbar\omega}) \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \text{Im} \tilde{\chi}_{abkl}^{\text{res}}(T, \omega) \int_0^\infty \frac{\text{Im} \tilde{\chi}_{cdij}^{\text{res}}(T, \Omega)}{\omega + \Omega} d\Omega \quad (\text{C.18})
\end{aligned}$$

where in the above calculation we insert the identity  $\sum_p |p\rangle \langle p|$ .



Term(3):

$$\begin{aligned}
& 2\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \sum_{ijkl} \sum_{ss'} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \sum_{ln} \langle m | \hat{T}_{ij}^{(s)} | n \rangle \frac{\langle n | \hat{V} | l \rangle}{E_l - E_n} \langle l | \hat{T}_{kl}^{(s')} | m \rangle \delta(\hbar\omega - E_l + E_m) \\
= & 4\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_{lnp} \frac{\delta(\hbar\omega - E_l + E_m)}{E_l - E_n} \langle m | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{ab}^{(s)} | p \rangle \langle p | \hat{T}_{cd}^{(s')} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | m \rangle \\
= & 4\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_{ln} \frac{\delta(\hbar\omega - E_l + E_m)}{E_l - E_n} \text{Tr} \left( \hat{T}_{cd}^{(s')} | l \rangle \langle l | \hat{T}_{kl}^{(s')} | m \rangle \langle m | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{ab}^{(s)} \right) \\
= & 4\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \\
& \sum_{l^{(s)} l^{(r)} l^{(s')} l^{(r')} n^{(s)} n^{(s')} n^{(r)}} \frac{\delta(\hbar\omega - E_l^{(s)} - E_l^{(s')} - \sum E_l^{(r)} + E_m^{(s)} + E_m^{(s')} + \sum E_m^{(r)})}{E_l^{(s)} + E_l^{(s')} + E_l^{(r)} - E_n^{(s)} - E_n^{(s')} - E_n^{(r)}} \\
& \text{Tr} \prod_{r \neq s, s'} \left( |l^{(r)}\rangle \langle l^{(r)}| m^{(r)} \rangle \langle m^{(r)}| n^{(r)} \rangle \langle n^{(r)}| \right) \\
& \text{Tr} \left( \hat{T}_{cd}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{kl}^{(s')} | m^{(s')} \rangle \langle m^{(s')} | n^{(s')} \rangle \langle n^{(s')} | \right) \text{Tr} \left( |l^{(s)}\rangle \langle l^{(s)}| m^{(s)} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ab}^{(s)} \right) \\
= & 4\pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_m \frac{e^{-\beta E_m}}{\mathcal{Z}} \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \sum_{l^{(s')} n^{(s)}} \frac{\delta(\hbar\omega - E_l^{(s')} + E_m^{(s')})}{\hbar\omega - E_n^{(s)}} \\
& \langle m^{(s')} | \hat{T}_{cd}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{kl}^{(s')} | m^{(s')} \rangle \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ab}^{(s)} | m^{(s)} \rangle \\
= & -4 \frac{L_0^3}{\pi} \pi\omega t (1 - e^{-\beta\hbar\omega}) \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \sum_{m^{(s')}} \frac{e^{-\beta E_m^{(s')}}}{\mathcal{Z}^{(s')}} \text{Im} \tilde{\chi}_{cdkl}^{(m^{(s')})}(\omega) \\
& \sum_{m^{(s)}} \frac{e^{-\beta E_m^{(s)}}}{\mathcal{Z}^{(s)}} \sum_{n^{(s)}} \frac{1}{\hbar\omega - E_n^{(s)}} \langle m^{(s)} | \hat{T}_{ij}^{(s)} | n^{(s)} \rangle \langle n^{(s)} | \hat{T}_{ab}^{(s)} | m^{(s)} \rangle \\
= & -4 \frac{L_0^6}{\pi} \omega t (1 - e^{-\beta\hbar\omega}) \sum_{ijklabcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} e_{ij} e_{kl} e^{-ik \cdot (r_s - r'_s)} \text{Im} \tilde{\chi}_{cdkl}^{\text{res}}(T, \omega) \int_0^\infty \frac{\text{Im} \tilde{\chi}_{ijab}^{\text{res}}(T, \Omega)}{\Omega^2 - \omega^2} d\Omega \quad (\text{C.19})
\end{aligned}$$

Finally we sum term(2-5) together. Their summation is given as follows, which turns out to be

$$\begin{aligned}
\dot{V}_{i,t}(L) &= \omega (1 - e^{-\beta\hbar\omega}) \frac{8N_0^3 L^3 A^2 k^2 \ln N_0}{\pi \rho c_{t,l}^2} \text{Im} \tilde{\chi}_{t,l}^{\text{res}}(T, \omega) \int_0^\infty \frac{\Omega \text{Im} \tilde{\chi}_{t,l}^{\text{res}}(T, \Omega)}{\Omega^2 - \omega^2} d\Omega \\
&= 4 (1 - e^{-\beta\hbar\omega}) \omega \frac{N_0^3 L^3 A^2 k^2 \ln N_0}{\rho c_{t,l}^2} \text{Im} \tilde{\chi}_{t,l}^{\text{res}}(T, \omega) \text{Re} \tilde{\chi}_{t,l}^{\text{res}}(T, \omega) \quad (\text{C.20})
\end{aligned}$$

where please note that the reduced imaginary part of resonance susceptibility is negative, and the reduced real part of resonance susceptibility is negative as well. Therefore the above result, Eq.(C.20) is positive.

This is Eq.(6.22) in chapter 6. In the above calculations we made use of the definition for “real part reduced resonance susceptibility”  $\text{Re } \tilde{\chi}_{l,t}^{\text{res}}(T, \omega)$ :

$$\text{Re } \tilde{\chi}_{l,t}^{\text{res}}(T, \omega) = \frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\Omega \text{Im } \tilde{\chi}_{l,t}^{\text{res}}(T, \Omega)}{\Omega^2 - \omega^2} d\Omega \quad (\text{C.21})$$

Also, the “imaginary part reduced resonance susceptibility” can be easily transformed back from that of real part:

$$\text{Im } \tilde{\chi}_{l,t}^{\text{res}}(T, \omega) = -\frac{2}{\pi} \mathcal{P} \int_0^\infty \frac{\omega \text{Re } \tilde{\chi}_{l,t}^{\text{res}}(T, \Omega)}{\Omega^2 - \omega^2} d\Omega \quad (\text{C.22})$$

This relation will be useful in deriving the Meissner-Berret ratio self-consistent equation.

## C.2.2 Calculation Details of Term[2]: Eq.(6.20, 6.21) in Chapter 6

Again, we are only interested in the quadratic order of non-elastic susceptibility. Because term[2] is the expectation value of operator of quadratic  $\delta\hat{V}$ , the leading order of term[2] is already in terms of quadratic non-elastic susceptibility. Thus we can use the approximations  $|n^*\rangle \rightarrow |n\rangle$ ,  $E_n^* \rightarrow E_n$  and  $\mathcal{Z}^* \rightarrow \mathcal{Z}$ .

$$\begin{aligned} & \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n^*}}{\mathcal{Z}^*} \int^t dt' dt'' \sum_{l^*} e^{-i(E_l^* - E_n^*)(t' - t'')/\hbar} (E_l^* - E_n^*) \langle n^* | \delta\hat{V}(t') | l^* \rangle \langle l^* | \delta\hat{V}(t'') | n^* \rangle \\ & \approx \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \int^t dt' dt'' \sum_l e^{-i(E_l - E_n)(t' - t'')/\hbar} (E_l - E_n) \langle n | \delta\hat{V}(t') | l \rangle \langle l | \delta\hat{V}(t'') | n \rangle \\ \text{term(6)} & = \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \int^t dt' dt'' \sum_l e^{-i(E_l - E_n)(t' - t'')/\hbar} (E_l - E_n) \\ & \quad \langle n | \sum_{s \neq s'} \sum_{ijkl}^{N_0^3} \left( \Delta\Lambda_{ijkl}^{(ss')} (t') \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \right) | l \rangle \langle l | \sum_{u \neq u'} \sum_{abcd}^{N_0^3} \left( \Delta\Lambda_{abcd}^{(uu')} (t'') \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} \right) | n \rangle \\ \text{term(7)} & + \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \int^t dt' dt'' \sum_l e^{-i(E_l - E_n)(t' - t'')/\hbar} (E_l - E_n) \\ & \quad \langle n | \sum_{s \neq s'} \sum_{ijkl}^{N_0^3} \left( 2\Lambda_{ijkl}^{(ss')} \Delta\hat{T}_{ij}^{(s)} (t') \hat{T}_{kl}^{(s')} \right) | l \rangle \langle l | \sum_{u \neq u'} \sum_{abcd}^{N_0^3} \left( 2\Lambda_{abcd}^{(uu')} \Delta\hat{T}_{ab}^{(u)} (t'') \hat{T}_{cd}^{(u')} \right) | n \rangle \\ \text{term(8)} & + \frac{2}{\hbar^2} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \int^t dt' dt'' \sum_l e^{-i(E_l - E_n)(t' - t'')/\hbar} (E_l - E_n) \\ & \quad \langle n | \sum_{s \neq s'} \sum_{ijkl}^{N_0^3} \left( \Delta\Lambda_{ijkl}^{(ss')} (t') \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \right) | l \rangle \langle l | \sum_{u \neq u'} \sum_{abcd}^{N_0^3} \left( 2\Lambda_{abcd}^{(uu')} \Delta\hat{T}_{ab}^{(u)} (t'') \hat{T}_{cd}^{(u')} \right) | n \rangle \end{aligned} \quad (\text{C.23})$$

Let's analyze them one by one. First of all we are able to calculate term(6). Before doing it's calculation let us first try to transform the matrix element product  $\sum_l \langle p | \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} | l \rangle \langle l | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | p \rangle \delta(\hbar\omega_l - \hbar\omega_p - \hbar\omega)$  into

the product of non-elastic susceptibilities:

$$\begin{aligned}
& \sum_l \langle p | \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} | l \rangle \langle l | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | p \rangle \delta(\hbar\omega_l - \hbar\omega_p - \hbar\omega) \\
&= \sum_l \sum_{nm} \langle p | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle \langle l | \hat{T}_{cd}^{(s')} | m \rangle \langle m | \hat{T}_{ab}^{(s)} | p \rangle \delta(\hbar\omega_l - \hbar\omega_p - \hbar\omega) \\
&= \sum_l \sum_{nm} \langle m | \hat{T}_{ab}^{(s)} | p \rangle \langle p | \hat{T}_{ij}^{(s)} | n \rangle \langle n | \hat{T}_{kl}^{(s')} | l \rangle \langle l | \hat{T}_{cd}^{(s')} | m \rangle \delta(\hbar\omega_l - \hbar\omega_p - \hbar\omega) \\
&= \sum_l \text{Tr} \left( \hat{T}_{ab}^{(s)} | p \rangle \langle p | \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} | l \rangle \langle l | \hat{T}_{cd}^{(s')} \right) \delta(\hbar\omega_l - \hbar\omega_p - \hbar\omega) \\
&= \sum_l \text{Tr} \left( |l^{(s)}\rangle \langle l^{(s)}| \hat{T}_{ab}^{(s)} |p^{(s)}\rangle \langle p^{(s)}| \hat{T}_{ij}^{(s)} \otimes |p^{(s')}\rangle \langle p^{(s')}| \hat{T}_{kl}^{(s')} |l^{(s')}\rangle \langle l^{(s')}| \hat{T}_{cd}^{(s')} \right. \\
&\quad \left. \otimes \prod_{r \neq s, s'} |p^{(r)}\rangle \langle p^{(r)}| \otimes \prod_{t \neq s, s'} |l^{(t)}\rangle \langle l^{(t)}| \right) \delta(\hbar\omega_l - \hbar\omega_p - \hbar\omega) \\
&= \sum_{l_s l'_s} \text{Tr} \left( |l^{(s)}\rangle \langle l^{(s)}| \hat{T}_{ab}^{(s)} |p^{(s)}\rangle \langle p^{(s)}| \hat{T}_{ij}^{(s)} \right) \text{Tr} \left( |p^{(s')}\rangle \langle p^{(s')}| \hat{T}_{kl}^{(s')} |l^{(s')}\rangle \langle l^{(s')}| \hat{T}_{cd}^{(s')} \right) \\
&\quad \text{Tr} \left( \prod_{r \neq s, s'} |p^{(r)}\rangle \langle p^{(r)}| \right) \text{Tr} \left( \prod_{t \neq s, s'} |l^{(t)}\rangle \langle l^{(t)}| \right) \delta(\hbar\omega_l - \hbar\omega_p - \hbar\omega) \\
&= \sum_{l_s l'_s} \langle l^{(s)} | \hat{T}_{ab}^{(s)} | p^{(s)} \rangle \langle p^{(s)} | \hat{T}_{ij}^{(s)} | l^{(s)} \rangle \langle p^{(s')} | \hat{T}_{kl}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{cd}^{(s')} | p^{(s')} \rangle \\
&\quad \delta(\hbar\omega_l^{(s)} + \hbar\omega_l^{(s')} - \hbar\omega_p^{(s)} - \hbar\omega_p^{(s')} - \hbar\omega) \\
&= \sum_{l_s} \left[ \int d(\hbar\omega_s) \langle p^{(s)} | \hat{T}_{ij}^{(s)} | l^{(s)} \rangle \langle l^{(s)} | \hat{T}_{ab}^{(s)} | p^{(s)} \rangle \delta(\hbar\omega_l^{(s)} - \hbar\omega_p^{(s)} - \hbar\omega_s) \right] \\
&\quad \sum_{l'_s} \left[ \int d(\hbar\omega'_s) \langle p^{(s')} | \hat{T}_{kl}^{(s')} | l^{(s')} \rangle \langle l^{(s')} | \hat{T}_{cd}^{(s')} | p^{(s')} \rangle \delta(\hbar\omega_l^{(s')} - \hbar\omega_p^{(s')} - \hbar\omega'_s) \right] \\
&\quad \delta(\hbar\omega_l^{(s)} + \hbar\omega_l^{(s')} - \hbar\omega_p^{(s)} - \hbar\omega_p^{(s')} - \hbar\omega) \\
&= \frac{L^6}{\pi^2} \int \text{Im} \tilde{\chi}_{ijab}^{(p_s)}(\omega_s) d(\hbar\omega_s) \int \text{Im} \tilde{\chi}_{cdkl}^{(p'_s)}(\omega'_s) d(\hbar\omega'_s) \delta(\hbar\omega_s + \hbar\omega'_s - \hbar\omega) \\
&= \frac{L^6}{\pi^2} \int \text{Im} \tilde{\chi}_{ijab}^{(p_s)}(\omega_s) \text{Im} \tilde{\chi}_{cdkl}^{(p'_s)}(\omega - \omega_s) d(\hbar\omega_s) \tag{C.24}
\end{aligned}$$

Since the phonon wave displacement  $\vec{u}(\vec{x}, t) = \vec{A} \cos(\omega t - k \cdot x)$ , we have  $\Delta x_{ss'} = |\vec{u}(\vec{x}, t) - \vec{u}(\vec{x}', t)| = A(k \cdot (x_s - x'_s)) |\sin(\omega t - k \cdot (x_s + x'_s)/2)|$ . So  $\Delta x_{ss'}/x_{ss'} = A \sum_i k_i n_i |\sin(\omega t - k \cdot (x_s + x'_s)/2)|$ . We can further denote  $\Delta \tilde{\Lambda}_{ijkl} = \lambda_{ijkl} |\sin(\omega t - k \cdot (x_s + x'_s)/2)|$ :

$$\begin{aligned}
\lambda_{ijkl}^{(ss')} &= \left\{ \frac{3}{4} \left[ 2(n_j n_l \delta_{ik} + n_j n_k \delta_{il} + n_i n_k \delta_{jl} + n_i n_l \delta_{jk}) \cos \theta_{ss'} \right. \right. \\
&\quad \left. \left. - [(m_j n_l + m_l n_j) \delta_{ik} + (m_j n_k + m_k n_j) \delta_{il} + (m_i n_k + m_k n_i) \delta_{jl} + (m_i n_l + m_l n_i)] \delta_{jk} \right] \right. \\
&\quad \left. - 3\alpha \cos \theta_{ss'} \left( n_k n_l \delta_{ij} + n_j n_l \delta_{ik} + n_k n_j \delta_{il} + n_i n_l \delta_{jk} + n_i n_k \delta_{jl} + n_i n_j \delta_{kl} \right) \right. \\
&\quad \left. + \frac{3}{2} \alpha \left[ m_i (n_l \delta_{jk} + n_k \delta_{jl} + n_j \delta_{kl}) + m_j (n_l \delta_{ik} + n_k \delta_{il} + n_i \delta_{kl}) \right. \right. \\
&\quad \left. \left. + m_k (n_l \delta_{ij} + n_i \delta_{jl} + n_j \delta_{il}) + m_l (n_k \delta_{ij} + n_i \delta_{jk} + n_j \delta_{ik}) \right] \right. \\
&\quad \left. - \frac{15}{2} \alpha \left( m_i n_j n_k n_l + m_j n_i n_k n_l + m_k n_i n_j n_l + m_l n_i n_j n_k \right) + 30\alpha n_i n_j n_k n_l \cos \theta_{ss'} \right\} A \vec{k} \cdot \vec{n} \quad (\text{C.25})
\end{aligned}$$

Hence term(6) can be simplified as

$$\begin{aligned}
\text{term(6)} &= \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \int^t dt' dt'' \sum_l e^{-i(E_l - E_n)(t' - t'')/\hbar} (E_l - E_n) \\
&\quad \langle n | \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \left( \Delta \Lambda_{ijkl}^{(ss')} (t') \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \right) | l \rangle \langle l | \sum_{u \neq u'}^{N_0^3} \sum_{abcd} \left( \Delta \Lambda_{abcd}^{(uu')} (t'') \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} \right) | n \rangle \\
&= \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \int^t dt' dt'' \sum_l e^{-i(E_l - E_n)(t' - t'')/\hbar} (E_l - E_n) \\
&\quad \langle n | \sum_{ss'} \sum_{ijkl} \left| \sin \left( \omega t - k \cdot \frac{x_s + x'_s}{2} \right) \right| \lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} | l \rangle \\
&\quad \langle l | \sum_{uu'} \sum_{abcd} \left| \sin \left( \omega t'' - k \cdot \frac{x_u + x'_u}{2} \right) \right| \lambda_{abcd}^{(uu')} \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} | n \rangle \\
&= \frac{\pi t \omega}{4} (1 - e^{-\beta \hbar \omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_l \sum_{ss'} \sum_{ijklabcd} \\
&\quad \lambda_{ijkl}^{(ss')} \lambda_{abcd}^{(ss')} \delta(\hbar \omega - E_l + E_n) \langle n | \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} | l \rangle \langle l | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} | n \rangle \\
&= \frac{\pi t \omega}{4} (1 - e^{-\beta \hbar \omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ss'} \sum_{ijklabcd} \\
&\quad \lambda_{ijkl}^{(ss')} \lambda_{abcd}^{(ss')} \frac{L^6}{\pi^2} \int \text{Im} \tilde{\chi}_{ijab}^{(n_s)}(\omega_s) \text{Im} \tilde{\chi}_{cdkl}^{(n'_s)}(\omega - \omega_s) d(\hbar \omega_s) \\
&= \frac{\pi t \omega}{4} (1 - e^{-\beta \hbar \omega}) \sum_{ss'} \sum_{ijklabcd} \lambda_{ijkl}^{(ss')} \lambda_{abcd}^{(ss')} \frac{L^6}{\pi^2} \int \text{Im} \tilde{\chi}_{ijab}^{\text{res}}(T, \omega_s) \text{Im} \tilde{\chi}_{cdkl}^{\text{res}}(T, \omega - \omega_s) d(\hbar \omega_s) \\
&= \hbar \omega t \frac{N_0^3 A^2 k^2 \ln N_0}{40 \pi^3 \rho^2 c_t^4} (1 - e^{-\beta \hbar \omega}) \int \text{Im} \tilde{\chi}_t^{\text{res}}(T, \Omega) \text{Im} \tilde{\chi}_t^{\text{res}}(T, \omega - \Omega) d\Omega \\
(\text{Longitudinal}) &\quad \times ((55 + 176\alpha + 688\alpha^2) + 44(1 + 4\alpha + 4\alpha^2)x(T, \omega)) \\
(\text{Transverse}) &\quad \times ((35 + 112\alpha + 656\alpha^2) + 28(1 + 4\alpha + 4\alpha^2)x(T, \omega)) \quad (\text{C.26})
\end{aligned}$$

where  $\alpha = 1 - c_t^2/c_l^2$  and  $x(T, \omega) = \chi_l(T, \omega)/\chi_t(T, \omega) - 2$ . This result appears as Eq.(6.20) in chapter 6.

I feel really exhausted here... anyway let's continue to write the details of obtaining Eq.(6.21) in chapter 6. Because we only qualitatively know the behavior of the change of stress tensor operator is  $\Delta\hat{T}_{ij} \sim e(t)\hat{T}_{ij}$ , we are only able to give a qualitative calculation to term(7) and term(8):

$$\begin{aligned}
\text{term(7)} & \quad \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \int^t dt' dt'' \sum_l e^{-i(E_l - E_n)(t' - t'')/\hbar} (E_l - E_n) \\
& \quad \langle n | \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \left( 2\Lambda_{ijkl}^{(ss')} \Delta\hat{T}_{ij}^{(s)}(t') \hat{T}_{kl}^{(s')} \right) |l\rangle \langle l| \sum_{u \neq u'}^{N_0^3} \sum_{abcd} \left( 2\Lambda_{abcd}^{(uu')} \Delta\hat{T}_{ab}^{(u)}(t'') \hat{T}_{cd}^{(u')} \right) |n\rangle \\
& \sim e(t') e(t'') \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \int^t dt' dt'' \sum_l e^{-i(E_l - E_n)(t' - t'')/\hbar} (E_l - E_n) \\
& \quad \langle n | \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \left( 2\Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \right) |l\rangle \langle l| \sum_{u \neq u'}^{N_0^3} \sum_{abcd} \left( 2\Lambda_{abcd}^{(uu')} \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} \right) |n\rangle \\
& = A^2 k^2 \pi t \omega (1 - e^{-\beta \hbar \omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_l \sum_{ss'} \sum_{ijklabcd} \\
& \quad \Lambda_{ijkl}^{(ss')} \Lambda_{abcd}^{(ss')} \delta(\hbar \omega - E_l + E_n) \langle n | \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} |l\rangle \langle l | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} |n\rangle \\
& = A^2 k^2 \pi t \omega (1 - e^{-\beta \hbar \omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ss'} \sum_{ijklabcd} \\
& \quad \Lambda_{ijkl}^{(ss')} \Lambda_{abcd}^{(ss')} \frac{L^6}{\pi^2} \int \text{Im} \tilde{\chi}_{ijab}^{(n_s)}(\omega_s) \text{Im} \tilde{\chi}_{cdkl}^{(n'_s)}(\omega - \omega_s) d(\hbar \omega_s) \\
& = A^2 k^2 \pi t \omega (1 - e^{-\beta \hbar \omega}) \sum_{ss'} \sum_{ijklabcd} \Lambda_{ijkl}^{(ss')} \Lambda_{abcd}^{(ss')} \frac{L^6}{\pi^2} \\
& \quad \int \text{Im} \tilde{\chi}_{ijab}^{\text{res}}(T, \omega_s) \text{Im} \tilde{\chi}_{cdkl}^{\text{res}}(T, \omega - \omega_s) d(\hbar \omega_s) \\
& \sim K_{l,t}^{(1)} \hbar \omega t \frac{N_0^3 A^2 k^2 \ln N_0}{\pi^3 \rho^2 c_t^4} (1 - e^{-\beta \hbar \omega}) \int \text{Im} \tilde{\chi}_t^{\text{res}}(T, \Omega) \text{Im} \tilde{\chi}_t^{\text{res}}(T, \omega - \Omega) d\Omega \quad (\text{C.27})
\end{aligned}$$

where  $K_{l,t}^{(1)}$  are constants for longitudinal and transverse cases, of order  $\sim 1$ . The calculation for term(8) is similar:

$$\begin{aligned}
\text{term(8)} & \frac{2}{\hbar^2} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \int^t dt' dt'' \sum_l e^{-i(E_l - E_n)(t' - t'')/\hbar} (E_l - E_n) \\
& \langle n | \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \left( \Delta \Lambda_{ijkl}^{(ss')} (t') \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \right) |l\rangle \langle l| \sum_{u \neq u'}^{N_0^3} \sum_{abcd} \left( 2\Lambda_{abcd}^{(uu')} \Delta \hat{T}_{ab}^{(u)} (t'') \hat{T}_{cd}^{(u')} \right) |n\rangle \\
& \sim \left| \sin \left( \omega t' - k \cdot \frac{x_s + x'_s}{2} \right) \right| e(t'') \frac{1}{\hbar^2} \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \int^t dt' dt'' \sum_l e^{-i(E_l - E_n)(t' - t'')/\hbar} (E_l - E_n) \\
& \langle n | \sum_{s \neq s'}^{N_0^3} \sum_{ijkl} \left( \lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \right) |l\rangle \langle l| \sum_{u \neq u'}^{N_0^3} \sum_{abcd} \left( 2\Lambda_{abcd}^{(uu')} \hat{T}_{ab}^{(u)} \hat{T}_{cd}^{(u')} \right) |n\rangle \\
& = \frac{1}{2} A^2 k^2 \pi t \omega (1 - e^{-\beta \hbar \omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_l \sum_{ss'} \sum_{ijklabcd} \\
& \lambda_{ijkl}^{(ss')} \Lambda_{abcd}^{(ss')} \delta(\hbar \omega - E_l + E_n) \langle n | \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} |l\rangle \langle l | \hat{T}_{ab}^{(s)} \hat{T}_{cd}^{(s')} |n\rangle \\
& = \frac{1}{2} A^2 k^2 \pi t \omega (1 - e^{-\beta \hbar \omega}) \sum_n \frac{e^{-\beta E_n}}{\mathcal{Z}} \sum_{ss'} \sum_{ijklabcd} \\
& \lambda_{ijkl}^{(ss')} \Lambda_{abcd}^{(ss')} \frac{L^6}{\pi^2} \int \text{Im} \tilde{\chi}_{ijab}^{(n_s)}(\omega_s) \text{Im} \tilde{\chi}_{cdkl}^{(n'_s)}(\omega - \omega_s) d(\hbar \omega_s) \\
& = \frac{1}{2} A^2 k^2 \pi t \omega (1 - e^{-\beta \hbar \omega}) \sum_{ss'} \sum_{ijklabcd} \lambda_{ijkl}^{(ss')} \Lambda_{abcd}^{(ss')} \frac{L^6}{\pi^2} \\
& \int \text{Im} \tilde{\chi}_{ijab}^{\text{res}}(T, \omega_s) \text{Im} \tilde{\chi}_{cdkl}^{\text{res}}(T, \omega - \omega_s) d(\hbar \omega_s) \\
& \sim K_{l,t}^{(2)} \hbar \omega t \frac{N_0^3 A^2 k^2 \ln N_0}{\pi^3 \rho^2 c_t^4} (1 - e^{-\beta \hbar \omega}) \int \text{Im} \tilde{\chi}_t^{\text{res}}(T, \Omega) \text{Im} \tilde{\chi}_t^{\text{res}}(T, \omega - \Omega) d\Omega \tag{C.28}
\end{aligned}$$

where  $K_{l,t}^{(2)}$  are constants for longitudinal and transverse cases, of order  $\sim 1$ . In Eq.(6.21), chapter 6 we define  $K_{l,t} = K_{l,t}^{(1)} + K_{l,t}^{(2)}$ .

## Appendix D

# The Details of the Matrix Form of the Inverse of Elastic Susceptibility

The elastic susceptibility is usually written as the form as follows.

$$\chi_{ijkl}^{\text{el}} = (\rho c_t^2 - 2\rho c_t^2)\delta_{ij}\delta_{kl} + \rho c_t^2(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) \quad (\text{D.1})$$

We want to reexpress it in the form of  $6 \times 6$  matrix representation, where we take the basis as  $(xx)$ ,  $(xy)$ ,  $(xz)$ ,  $(yy)$ ,  $(yz)$ ,  $(zz)$ . In the following discussions we use 1, 2, 3 to represent  $x, y, z$ . To calculate the matrix form of elastic susceptibility, we let the double indices  $(ij)$  and  $(kl)$  in elastic susceptibility  $\chi_{ijkl}^{\text{el}}$  to take  $(ij)$  and  $(kl)$  equal to (11), (12), (13), (22), (23), (33) and obtain the 36 matrix elements of  $\chi_{ijkl}^{\text{el}}$ :

$$\chi^{\text{el}} = \begin{pmatrix} \chi_{1111}^{\text{el}} & \chi_{1112}^{\text{el}} & \chi_{1113}^{\text{el}} & \chi_{1122}^{\text{el}} & \chi_{1123}^{\text{el}} & \chi_{1133}^{\text{el}} \\ \chi_{1211}^{\text{el}} & \chi_{1212}^{\text{el}} & \chi_{1213}^{\text{el}} & \chi_{1222}^{\text{el}} & \chi_{1223}^{\text{el}} & \chi_{1233}^{\text{el}} \\ \chi_{1311}^{\text{el}} & \chi_{1312}^{\text{el}} & \chi_{1313}^{\text{el}} & \chi_{1322}^{\text{el}} & \chi_{1323}^{\text{el}} & \chi_{1333}^{\text{el}} \\ \chi_{2211}^{\text{el}} & \chi_{2212}^{\text{el}} & \chi_{2213}^{\text{el}} & \chi_{2222}^{\text{el}} & \chi_{2223}^{\text{el}} & \chi_{2233}^{\text{el}} \\ \chi_{2311}^{\text{el}} & \chi_{2312}^{\text{el}} & \chi_{2313}^{\text{el}} & \chi_{2322}^{\text{el}} & \chi_{2323}^{\text{el}} & \chi_{2333}^{\text{el}} \\ \chi_{3311}^{\text{el}} & \chi_{3312}^{\text{el}} & \chi_{3313}^{\text{el}} & \chi_{3322}^{\text{el}} & \chi_{3323}^{\text{el}} & \chi_{3333}^{\text{el}} \end{pmatrix} \quad (\text{D.2})$$

We can calculate every of these matrix elements by putting indices  $ijkl$  into Eq.(D.1). The inverse of the above Eq.(D.2) is actually the  $6 \times 6$  matrix form of the inverse of elastic susceptibility, and it is further given as follows,

$$(\chi^{\text{el}})^{-1} = \frac{1}{\rho c_t^2} \begin{pmatrix} \frac{\alpha}{4\alpha-1} & 0 & 0 & -\frac{2\alpha-1}{2(4\alpha-1)} & 0 & -\frac{2\alpha-1}{2(4\alpha-1)} \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ -\frac{2\alpha-1}{2(4\alpha-1)} & 0 & 0 & \frac{\alpha}{4\alpha-1} & 0 & -\frac{2\alpha-1}{2(4\alpha-1)} \\ 0 & 0 & 0 & 0 & 1 & 0 \\ -\frac{2\alpha-1}{2(4\alpha-1)} & 0 & 0 & -\frac{2\alpha-1}{2(4\alpha-1)} & 0 & \frac{\alpha}{4\alpha-1} \end{pmatrix} \quad (\text{D.3})$$

## Appendix E

# Details of Calculations of Sound Velocity Shift as the Function of Logarithmic of Temperature

In this chapter we want to give a detailed calculation of Eq.(4.26) in chapter 4, with the assumption that the reduced imaginary resonance susceptibility  $\text{Im } \tilde{\chi}_{l,t}^{\text{res}}(\omega, T) = (1 - e^{-\beta\hbar\omega})^{-1} \text{Im } \chi_{l,t}^{\text{res}}(\omega, T)$  is approximately a constant of frequency and temperature up to  $\omega_c \sim 10^{15}$ Hz and around the temperature of order 1K[34, 25]. We write Eq.(4.26) in the following,

$$\left. \frac{\Delta c_{l,t}(T) - \Delta c_{l,t}(T_0)}{c_{l,t}(T_0)} \right|_{\text{res}} = \frac{2}{2\pi\rho c_{l,t}^2} \mathcal{P} \int_0^\infty \frac{\Omega \left( \text{Im } \chi_{l,t}^{\text{res}}(\Omega, T) - \text{Im } \chi_{l,t}^{\text{res}}(\Omega, T_0) \right)}{\Omega^2 - \omega^2} d\Omega = C_{l,t} \ln \left( \frac{T}{T_0} \right) \quad (\text{E.1})$$

Let us use the reduced imaginary resonance susceptibility  $\text{Im } \tilde{\chi}_{l,t}^{\text{res}}(\omega, T) \approx \text{Im } \tilde{\chi}_{l,t}^{\text{res}}$  in the above integral,

$$\begin{aligned} & \frac{1}{\pi\rho c_{l,t}^2} \mathcal{P} \int_0^\infty \frac{\Omega \left( \text{Im } \chi_{l,t}^{\text{res}}(\Omega, T) - \text{Im } \chi_{l,t}^{\text{res}}(\Omega, T_0) \right)}{\Omega^2 - \omega^2} d\Omega \\ &= \frac{\text{Im } \tilde{\chi}_{l,t}^{\text{res}}}{\pi\rho c_{l,t}^2} \mathcal{P} \int_0^\infty \frac{\Omega}{\Omega^2 - \omega^2} \left[ (1 - e^{-\beta\hbar\Omega}) - (1 - e^{-\beta_0\hbar\Omega}) \right] d\Omega \\ &= \frac{\text{Im } \tilde{\chi}_{l,t}^{\text{res}}}{\pi\rho c_{l,t}^2} \mathcal{P} \int_0^\infty \frac{\Omega}{\Omega^2 - \omega^2} (e^{-\beta_0\hbar\Omega} - e^{-\beta\hbar\Omega}) d\Omega \end{aligned} \quad (\text{E.2})$$

where we define  $\beta_0 = (k_B T_0)^{-1}$  and take the “frequency and temperature independent quantity”  $\text{Im } \tilde{\chi}_{l,t}^{\text{res}}$  out of the integral. Such frequency and temperature independence of  $\text{Im } \tilde{\chi}_{l,t}^{\text{res}}(\omega, T)$  is observed in experiments[34, 33]. Also, according to the argument by D. C. Vural and A. J. Leggett[25], the frequency dependence of imaginary part of non-elastic susceptibility  $\text{Im } \chi_{l,t}^{\text{res}}(\omega)$  does not differ that much for  $\tanh(\omega/2k_B T)$  function dependence (derived from TTLS model) and  $(1 - e^{-\beta\hbar\omega})$  function dependence (derived by multiple-level-system model). Therefore one would intuitive expect that the “logarithmic temperature dependence of sound velocity shift” should also be proven in arbitrary multiple-level-system similar with TTLS results. Please note the above integral Eq.(E.2) has many nice properties: (1) it converges exponentially fast with the increase of frequency variable  $\Omega$ ; (2) the “principle value” removes the divergence when  $\Omega$  approaches  $\omega$ . We will evaluate this principle integral in details as follows.

The ultrasonic sound velocity shift experiments are measured around the temperatures of order 1K, which



means the input ultrasonic phonon energy  $\hbar\omega \sim 10^{-28}\text{J}$  is much smaller than  $k_B T \sim 10^{-23}\text{J}$ . Thus when the integral variable  $\Omega$  approaches singularity  $\omega$ , the function  $e^{-\beta\hbar\Omega} \approx 1$ . The following principle integral

$$\begin{aligned} \mathcal{P} \int_0^{\omega_>} \frac{\Omega}{\Omega^2 - \omega^2} e^{-\beta\hbar\Omega} d\Omega &= \lim_{\epsilon \rightarrow 0} \left( \int_0^{\omega - \epsilon} \frac{\Omega}{\Omega^2 - \omega^2} e^{-\beta\hbar\Omega} d\Omega + \int_{\omega + \epsilon}^{\omega_>} \frac{\Omega}{\Omega^2 - \omega^2} e^{-\beta\hbar\Omega} d\Omega \right) \\ &= \frac{1}{2} \lim_{\epsilon \rightarrow 0} \left( \ln \frac{\epsilon}{\omega} + \ln \frac{\omega_>}{\epsilon} \right) = \frac{1}{2} \ln \left( \frac{\omega_>}{\omega} \right) \end{aligned} \quad (\text{E.3})$$

where  $\omega_>$  is “some” integral upper cut-off. From the exponentially decay behavior of function  $e^{-\beta\hbar\Omega}$ , we know this upper cut-off is some constant times temperature  $T$ :  $\omega_>(T) \propto T$ . Back to Eq.(E.2) we need to calculate both of the two parts in the bracket, it turns out to be

$$\begin{aligned} \left. \frac{\Delta c_{l,t}(T) - \Delta c_{l,t}(T_0)}{c_{l,t}(T_0)} \right|_{\text{res}} &= \frac{\text{Im} \tilde{\chi}_{l,t}^{\text{res}}}{\pi \rho c_{l,t}^2} \mathcal{P} \int_0^{\infty} \frac{\Omega}{\Omega^2 - \omega^2} (e^{-\beta_0 \hbar \Omega} - e^{-\beta \hbar \Omega}) d\Omega \\ &= \frac{\text{Im} \tilde{\chi}_{l,t}^{\text{res}}}{2\pi \rho c_{l,t}^2} \left[ \ln \left( \frac{\omega_>(T_0)}{\omega} \right) - \ln \left( \frac{\omega_>(T)}{\omega} \right) \right] \\ &= -\frac{\text{Im} \tilde{\chi}_{l,t}^{\text{res}}}{2\pi \rho c_{l,t}^2} \ln \left( \frac{\omega_>(T)}{\omega_>(T_0)} \right) = -\frac{\text{Im} \tilde{\chi}_{l,t}^{\text{res}}}{2\pi \rho c_{l,t}^2} \ln \left( \frac{T}{T_0} \right) \end{aligned} \quad (\text{E.4})$$

this is the details of Eq.(4.26) calculations, where  $-\frac{\text{Im} \tilde{\chi}_{l,t}^{\text{res}}}{2\pi \rho c_{l,t}^2}$  is the constant  $\mathcal{C}_{l,t}$  which appears in Eq.(4.26).

## Appendix F

# Calculation Details of the Coefficient Renormalization Equations (4.18)

In the renormalization eqatons, Eqs.(4.18),

$$\begin{aligned} \chi_{ijkl}^{\text{super}}(\omega) &= \frac{1}{1-i\omega\tau} \left\{ \chi_{ijkl}^{\text{rel}} - \frac{L_n^3}{N_0^3} \left[ -\sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} (0) e^{-i\vec{k}\cdot(\vec{x}_s-\vec{x}'_s)} \right] (\chi_{ijab}^{\text{rel}} \chi_{cdkl}^{\text{rel}} + 2\chi_{ijab}^{\text{rel}} \chi_{cdkl}^{\text{res}}(0)) \right\} \\ &+ \chi_{ijkl}^{\text{res}}(\omega+i\eta) - \frac{L_n^3}{N_0^3} \left[ -\sum_{abcd} \sum_{ss'} \Lambda_{abcd}^{(ss')} (0) e^{-i\vec{k}\cdot(\vec{x}_s-\vec{x}'_s)} \right] \chi_{ijab}^{\text{res}}(\omega+i\eta) \chi_{cdkl}^{\text{res}}(\omega+i\eta) \end{aligned} \quad (\text{F.1})$$

We are required to calculate the linear term expansion  $\sum_{abcd} \chi_{ijab} \frac{L_n^3}{N_0^3} \left[ -\sum_{ss'} \Lambda_{abcd}^{(ss')} (0) e^{-i\vec{k}\cdot(\vec{x}_s-\vec{x}'_s)} \right] \chi_{cdkl}$ , and simplify it into the following renormalization equations,

$$\begin{aligned} \chi_{t,l}^{\text{super rel}} &= \chi_{t,l}^{\text{rel}} - \frac{1}{\rho c_{t,l}^2} \left[ (\chi_{t,l}^{\text{rel}})^2 + 2\chi_{t,l}^{\text{rel}} \chi_{t,l}^{\text{res}}(0) \right] \\ \chi_{t,l}^{\text{super res}}(\omega+i\eta) &= \chi_{t,l}^{\text{res}}(\omega+i\eta) - \frac{1}{\rho c_{t,l}^2} \left[ \chi_{t,l}^{\text{res}}(\omega+i\eta) \right]^2 \end{aligned} \quad (\text{F.2})$$

First of all, let us give a short review of the process of calculating  $\Lambda_{abcd}^{(ss')}$ : the non-elastic stress-stress interaction is defined as follows,

$$\hat{V} = \sum_{s \neq s'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} \hat{T}_{ij}^{(s)} \hat{T}_{kl}^{(s')} \quad (\text{F.3})$$

where the coefficient  $\Lambda_{ijkl}^{(ss')}$  is the term  $\Lambda_{abcd}^{(ss')}$  in Eq.(F.1). In Appendix (A) the non-elastic stress-stress interaction is obtained by ‘‘completing the square of phonon Hamiltonian’’, and it is given as follows,

$$\begin{aligned} \hat{V} &= \frac{1}{2Nm} \left( \frac{1}{c_t^2} - \frac{1}{c_l^2} \right) \sum_{s \neq s'} \sum_{ijkl} \sum_{\vec{k}} \left( \frac{k_i k_j k_k k_l}{k^4} \right) \cos(\vec{k} \cdot \vec{x}_{ss'}) T_{ij}^{(s)} T_{kl}^{(s')} \\ &- \frac{1}{8Nm} \frac{1}{c_t^2} \sum_{s \neq s'} \sum_{ijkl} \sum_{\vec{k}} \left( \frac{k_j k_l \delta_{ik} + k_j k_k \delta_{il} + k_i k_l \delta_{jk} + k_i k_k \delta_{jl}}{k^2} \right) \cos(\vec{k} \cdot \vec{x}_{ss'}) T_{ij}^{(s)} T_{kl}^{(s')} \end{aligned} \quad (\text{F.4})$$

where  $\vec{x}_{ss'} = \vec{x}_s - \vec{x}'_s$ . Since we have the relation  $\cos(\vec{k} \cdot \vec{x}) = \frac{1}{2} (e^{-i\vec{k} \cdot \vec{x}} + e^{i\vec{k} \cdot \vec{x}})$ , the above Eq.(F.4) can be rewritten as

$$\begin{aligned} \hat{V} &= \frac{1}{2Nm} \left( \frac{1}{c_t^2} - \frac{1}{c_l^2} \right) \sum_{s \neq s'} \sum_{ijkl} \sum_{\vec{k}} \left( \frac{k_i k_j k_k k_l}{k^4} \right) \frac{1}{2} \left( e^{-i\vec{k} \cdot \vec{x}_{ss'}} + e^{i\vec{k} \cdot \vec{x}_{ss'}} \right) T_{ij}^{(s)} T_{kl}^{(s')} \\ &\quad - \frac{1}{8Nm} \frac{1}{c_t^2} \sum_{s \neq s'} \sum_{ijkl} \sum_{\vec{k}} \left( \frac{k_j k_l \delta_{ik} + k_j k_k \delta_{il} + k_i k_l \delta_{jk} + k_i k_k \delta_{jl}}{k^2} \right) \frac{1}{2} \left( e^{-i\vec{k} \cdot \vec{x}_{ss'}} + e^{i\vec{k} \cdot \vec{x}_{ss'}} \right) T_{ij}^{(s)} T_{kl}^{(s')} \end{aligned} \quad (\text{F.5})$$

Now, let us compare the following two pairs of summations:

$$\begin{aligned} &\frac{1}{2} \sum_{\vec{k}} \left( \frac{k_i k_j k_k k_l}{k^4} \right) e^{-i\vec{k} \cdot \vec{x}_{ss'}} \\ &\frac{1}{2} \sum_{\vec{k}} \left( \frac{k_i k_j k_k k_l}{k^4} \right) e^{+i\vec{k} \cdot \vec{x}_{ss'}} \\ &\frac{1}{2} \sum_{\vec{k}} \left( \frac{k_j k_l \delta_{ik} + k_j k_k \delta_{il} + k_i k_l \delta_{jk} + k_i k_k \delta_{jl}}{k^2} \right) e^{-i\vec{k} \cdot \vec{x}_{ss'}} \\ &\frac{1}{2} \sum_{\vec{k}} \left( \frac{k_j k_l \delta_{ik} + k_j k_k \delta_{il} + k_i k_l \delta_{jk} + k_i k_k \delta_{jl}}{k^2} \right) e^{+i\vec{k} \cdot \vec{x}_{ss'}} \end{aligned} \quad (\text{F.6})$$

please note, that in the above summations over momentum  $\vec{k}$ , the components  $k_i, k_j, k_k, k_l$  are the  $x, y, z$  components of  $\vec{k}$  when  $i, j, k, l$  equal to  $x, y, z$ . Therefore, the first pair of summations are equal to each other, and the second pair of summations are equal to each other as well. We prove these equal relations as follows. In the summation  $\frac{1}{2} \sum_{\vec{k}} \left( \frac{k_i k_j k_k k_l}{k^4} \right) e^{+i\vec{k} \cdot \vec{x}_{ss'}}$  we define  $\vec{k}' = -\vec{k}$ . Thus the summation is given by

$$\begin{aligned} \frac{1}{2} \sum_{\vec{k}} \left( \frac{k_i k_j k_k k_l}{k^4} \right) e^{+i\vec{k} \cdot \vec{x}_{ss'}} &= \frac{1}{2} \sum_{-\vec{k}'} \frac{(-k'_i)(-k'_j)(-k'_k)(-k'_l)}{k'^4} e^{-i\vec{k}' \cdot \vec{x}_{ss'}} \\ &= \frac{1}{2} \sum_{\vec{k}'} \left( \frac{k'_i k'_j k'_k k'_l}{k'^4} \right) e^{-i\vec{k}' \cdot \vec{x}_{ss'}} \\ &= \frac{1}{2} \sum_{\vec{k}} \left( \frac{k_i k_j k_k k_l}{k^4} \right) e^{-i\vec{k} \cdot \vec{x}_{ss'}} \end{aligned} \quad (\text{F.7})$$

In the summation  $\frac{1}{2} \sum_{\vec{k}} \left( \frac{k_j k_l \delta_{ik} + k_j k_k \delta_{il} + k_i k_l \delta_{jk} + k_i k_k \delta_{jl}}{k^2} \right) e^{+i\vec{k} \cdot \vec{x}_{ss'}}$  we define  $\vec{k}' = -\vec{k}$  as well. The summation is simplified as:

$$\begin{aligned}
& \frac{1}{2} \sum_{\vec{k}} \left( \frac{k_j k_l \delta_{ik} + k_j k_k \delta_{il} + k_i k_l \delta_{jk} + k_i k_k \delta_{jl}}{k^2} \right) e^{+i\vec{k} \cdot \vec{x}_{ss'}} \\
&= \frac{1}{2} \sum_{-\vec{k}'} \left( \frac{(-k'_j)(-k'_l) \delta_{ik} + (-k'_j)(-k'_k) \delta_{il} + (-k'_i)(-k'_l) \delta_{jk} + (-k'_i)(-k'_k) \delta_{jl}}{k'^2} \right) e^{-i\vec{k}' \cdot \vec{x}_{ss'}} \\
&= \frac{1}{2} \sum_{-\vec{k}'} \left( \frac{k'_j k'_l \delta_{ik} + k'_j k'_k \delta_{il} + k'_i k'_l \delta_{jk} + k'_i k'_k \delta_{jl}}{k'^2} \right) e^{-i\vec{k}' \cdot \vec{x}_{ss'}} \\
&= \frac{1}{2} \sum_{\vec{k}'} \left( \frac{k'_j k'_l \delta_{ik} + k'_j k'_k \delta_{il} + k'_i k'_l \delta_{jk} + k'_i k'_k \delta_{jl}}{k'^2} \right) e^{-i\vec{k}' \cdot \vec{x}_{ss'}} \\
&= \frac{1}{2} \sum_{\vec{k}} \left( \frac{k_j k_l \delta_{ik} + k_j k_k \delta_{il} + k_i k_l \delta_{jk} + k_i k_k \delta_{jl}}{k^2} \right) e^{-i\vec{k} \cdot \vec{x}_{ss'}} \tag{F.8}
\end{aligned}$$

Therefore the non-elastic stress-stress interaction can be rewritten as

$$\begin{aligned}
\hat{V} &= \sum_{s \neq s'} \sum_{ijkl} \Lambda_{ijkl}^{(ss')} T_{ij}^{(s)} T_{kl}^{(s')} \\
&= \frac{1}{2Nm} \left( \frac{1}{c_t^2} - \frac{1}{c_l^2} \right) \sum_{s \neq s'} \sum_{ijkl} \sum_{\vec{k}} \left( \frac{k_i k_j k_k k_l}{k^4} \right) e^{i\vec{k} \cdot \vec{x}_{ss'}} T_{ij}^{(s)} T_{kl}^{(s')} \\
&\quad - \frac{1}{8Nm} \frac{1}{c_t^2} \sum_{s \neq s'} \sum_{ijkl} \sum_{\vec{k}} \left( \frac{k_j k_l \delta_{ik} + k_j k_k \delta_{il} + k_i k_l \delta_{jk} + k_i k_k \delta_{jl}}{k^2} \right) e^{i\vec{k} \cdot \vec{x}_{ss'}} T_{ij}^{(s)} T_{kl}^{(s')} \tag{F.9}
\end{aligned}$$

where

$$\begin{aligned}
\Lambda_{ijkl}^{(ss')} &= \frac{1}{2Nm} \left( \frac{1}{c_t^2} - \frac{1}{c_l^2} \right) \sum_{\vec{k}} \left( \frac{k_i k_j k_k k_l}{k^4} \right) e^{i\vec{k} \cdot \vec{x}_{ss'}} \\
&\quad - \frac{1}{8Nm} \frac{1}{c_t^2} \sum_{\vec{k}} \left( \frac{k_j k_l \delta_{ik} + k_j k_k \delta_{il} + k_i k_l \delta_{jk} + k_i k_k \delta_{jl}}{k^2} \right) e^{i\vec{k} \cdot \vec{x}_{ss'}} \tag{F.10}
\end{aligned}$$

The above Eq.(F.10) is the most original definition of  $\Lambda_{ijkl}^{(ss')}$ . Now let us begin to calculate the linear term  $\sum_{abcd} \chi_{ijab} \frac{L_n^3}{N_0^3} \left[ -\sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-i\vec{k} \cdot (x_s - x'_s)} \right] \chi_{cdkl}$ . For notation simplicity let us denote

$$f_{ijkl}(\vec{k}) = \frac{1}{2Nm} \left( \frac{1}{c_t^2} - \frac{1}{c_l^2} \right) \left( \frac{k_i k_j k_k k_l}{k^4} \right) - \frac{1}{8Nm} \frac{1}{c_t^2} \left( \frac{k_j k_l \delta_{ik} + k_j k_k \delta_{il} + k_i k_l \delta_{jk} + k_i k_k \delta_{jl}}{k^2} \right) \tag{F.11}$$

So  $\Lambda_{ijkl}^{(ss')} = \sum_{\vec{k}} f_{ijkl}(\vec{k}) e^{i\vec{k} \cdot (\vec{x}_s - \vec{x}'_s)}$ . The linear term  $\sum_{abcd} \chi_{ijab} \frac{L_n^3}{N_0^3} \left[ -\sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-i\vec{k} \cdot (x_s - x'_s)} \right] \chi_{cdkl}$  is calculated as follows,

$$\begin{aligned}
& \sum_{abcd} \chi_{ijab} \frac{L_n^3}{N_0^3} \left[ - \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \right] \chi_{cdkl} \\
&= \sum_{abcd} \chi_{ijab} \frac{1}{8} \frac{L_n^3}{N_0^3} \left[ - \sum_{(\vec{x}_s + \vec{x}'_s)} \sum_{(\vec{x}_s - \vec{x}'_s)} \Lambda_{abcd}^{(ss')} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \right] \chi_{cdkl} \\
&= \sum_{abcd} \chi_{ijab} \frac{1}{8} \frac{L_n^3}{N_0^3} \left[ - \sum_{(\vec{x}_s + \vec{x}'_s)} \sum_{(\vec{x}_s - \vec{x}'_s)} \sum_{\vec{p}} f_{abcd}(\vec{p}) e^{i\vec{p}\cdot(\vec{x}_s - \vec{x}'_s)} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \right] \chi_{cdkl} \\
&= \sum_{abcd} \chi_{ijab} L_n^3 \left[ - \sum_{\vec{p}} f_{abcd}(\vec{p}) \sum_{(\vec{x}_s - \vec{x}'_s)} e^{i\vec{p}\cdot(\vec{x}_s - \vec{x}'_s)} e^{-i\vec{k}\cdot(\vec{x}_s - \vec{x}'_s)} \right] \chi_{cdkl} \\
&= \sum_{abcd} \chi_{ijab} L_n^3 \left[ - \sum_{\vec{p}} f_{abcd}(\vec{p}) \delta_{\vec{p}, \vec{k}} \right] \chi_{cdkl} \\
&= \sum_{abcd} \chi_{ijab} L_n^3 \left[ - f_{abcd}(\vec{k}) \right] \chi_{cdkl} \\
&= - \sum_{abcd} \chi_{ijab} \left[ \frac{1}{2\rho} \left( \frac{1}{c_t^2} - \frac{1}{c_l^2} \right) \left( \frac{k_a k_b k_c k_d}{k^4} \right) - \frac{1}{8\rho} \frac{1}{c_t^2} \left( \frac{k_b k_d \delta_{ac} + k_b k_c \delta_{ad} + k_a k_d \delta_{bc} + k_a k_c \delta_{bd}}{k^2} \right) \right] \chi_{cdkl} \\
&= \frac{1}{2\rho c_t^2} \sum_{abcd} \chi_{ijab} \left[ -\alpha \left( \frac{k_a k_b k_c k_d}{k^4} \right) + \frac{1}{4} \left( \frac{k_b k_d \delta_{ac} + k_b k_c \delta_{ad} + k_a k_d \delta_{bc} + k_a k_c \delta_{bd}}{k^2} \right) \right] \chi_{cdkl} \quad (\text{F.12})
\end{aligned}$$

where  $\alpha = 1 - c_t^2/c_l^2$ . Next we use the assumption, that single block susceptibility  $\lim_{\vec{k} \rightarrow 0} \chi_{ijkl}(\vec{k})$  should approximately be independent of the direction of momentum  $\vec{k}/k$  at small wave number limit  $\lim_{\vec{k} \rightarrow 0}$ . Therefore, we assume that the single block susceptibility take the generic isotropic form,  $\chi_{ijkl} = (\chi_l - 2\chi_t)\delta_{ij}\delta_{kl} + \chi_t(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$ . Let us define a new quantity, the ‘‘change of non-elastic susceptibility from single block to super block,  $\delta\chi_{ijkl} = \chi_{ijkl}^{\text{super}} - \chi_{ijkl}$ ’’.

In the renormalization equations, the ‘‘change of non-elastic susceptibility from single block to super block’’ is equal to Eq.(F.12). In Eq.(F.12) both of ‘‘ $\chi_{ijab}$ ’’ and ‘‘ $\chi_{cdkl}$ ’’ are independent of the direction of momentum, ‘‘ $\vec{k}/k$ ’’ at small wave number limit. After the summation over indices  $\sum_{abcd}$ , the quantity  $\frac{1}{2\rho c_t^2} \sum_{abcd} \chi_{ijab} \left[ -\alpha \left( \frac{k_a k_b k_c k_d}{k^4} \right) + \frac{1}{4} \left( \frac{k_b k_d \delta_{ac} + k_b k_c \delta_{ad} + k_a k_d \delta_{bc} + k_a k_c \delta_{bd}}{k^2} \right) \right] \chi_{cdkl}$  is independent of the direction of momentum as well.

Therefore, the terms with odd orders of momentum components  $k_x, k_y, k_z$  must vanish (because if we reverse the direction of momentum  $\vec{k}$ , the signs of odd terms are reversed); for even orders of momentum components  $k_x, k_y, k_z$ , they take their average values (because if we change the direction of momentum from  $\vec{k}$  to  $\vec{k}'$ , the ‘‘change of non-elastic susceptibility  $\delta\chi_{ijkl}$ ’’ in small wave number limit is independent of such

momentum direction change). For example,

$$\overline{k_a k_b} = \frac{\int \frac{d^3 k}{(2\pi)^3} k_a k_b}{\int \frac{d^3 k}{(2\pi)^3} k^2} = \frac{1}{3} \delta_{ab} \quad \overline{k_a k_b k_c k_d} = \frac{\int \frac{d^3 k}{(2\pi)^3} k_a k_b k_c k_d}{\int \frac{d^3 k}{(2\pi)^3} k^4} = \frac{1}{15} (\delta_{ab} \delta_{cd} + \delta_{ac} \delta_{bd} + \delta_{ad} \delta_{bc}) \quad (\text{F.13})$$

Finally we plug Eq.(F.13) into Eq.(F.12), and sum over the 81 summations to obtain the following result. This result implies that “the change of non-elastic susceptibility  $\delta\chi_{ijkl}$  takes the generic isotropic form in small wave number limit  $\lim_{\vec{k} \rightarrow 0}$ :  $\delta\chi_{ijkl} = (\delta\chi_l - 2\delta\chi_t)\delta_{ij}\delta_{kl} + \delta\chi_t(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$ ”,

$$\delta\chi_{ijkl} = \sum_{abcd} \chi_{ijab} \frac{L_n^3}{N_0^3} \left[ - \sum_{ss'} \Lambda_{abcd}^{(ss')} e^{-i\vec{k} \cdot (\vec{x}_s - \vec{x}'_s)} \right] \chi_{cdkl} = \left( \frac{\chi_l^2}{\rho c_l^2} - 2 \frac{\chi_t^2}{\rho c_t^2} \right) \delta_{ij} \delta_{kl} + \frac{\chi_t^2}{\rho c_t^2} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) \quad (\text{F.14})$$

which gives the simplified RG equations Eqs.(F.2).

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