

Thermal conductivity of glassy materials and the “boson peak”

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A theory for the anomalous vibrational and thermal properties of disordered solids based on the model assumption of randomly fluctuating transverse elastic constants is presented. Mean-field expressions for the vibrational density of states and the energy diffusivity are derived with field theoretical techniques. As in previous approaches of this type the boson peak (enhancement of the low-frequency density of states) is explained as a result of the frozen-in disorder and compares well with the experimental findings. The plateau in the temperature variation of the thermal conductivity and the behavior beyond the plateau is shown to arise from the enhanced scattering in the boson peak regime and to be essentially a harmonic phenomenon.

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The temperature dependence of the thermal conductivity $\kappa(T)$ of disordered solids such as glasses or strongly perturbed crystals is very different from that of the corresponding crystalline materials [1–8]. The almost quadratic variation of $\kappa(T)$ with temperature (as well as the almost linear one of the specific heat) around and below 1 K is usually explained in terms of inelastic scattering from two-level defects [9]. The present work focuses on the behavior around ~ 10 K (or $\sim 1/10$ of the Debye temperature θ_D). In this regime $\kappa(T)$ exhibits a plateau, followed by an increase and a final saturation. In the same temperature regime the “reduced specific heat” $C(T)/T^3$ exhibits a peak that can be traced to a peak in the “reduced density of states” (DOS) $g(\omega)/\omega^2$ (“boson peak”, BP).

The nature of the vibrational states near and above the BP frequency has been the subject-matter of a very intense and controversial debate among workers who performed neutron-, X-ray, and other inelastic scattering experiments [10–14], computer simulations [15–20], and analytical theory [21–29]. Although there is not yet a general consensus in the community most investigations show that these states are of harmonic nature and are strongly affected by the quenched disorder of the material of interest. From most of the quoted literature it emerges that the states near the BP and above are *not* localized, i.e. collective sound-like excitations with linear dispersion. However, they are of *diffusing, strong-scattering* character, which means that the width of the Brillouin-like maximum is of the same order as its principal frequency. These features, observed in experiments and simulations have been accounted for by most of the quoted mean-field approaches [23–29], in particular by treatments based on the assumption of random force constants [23, 25, 26, 28] or, as in our recent field-theoretical work [27], on random elastic constants. This picture is at variance with early models for $\kappa(T)$ near and beyond the plateau [30–33], in which the pertinent vibrational states were supposed to be localized. The increase of $\kappa(T)$ above 10 K was attributed to an anharmonic mechanism. In particular it has been shown by simulations

of thermal transport in glassy Si and SiO₂ [15, 19] that $\kappa(T)$ in this regime can be explained in terms of harmonic excitations.

The aim of the present contribution is twofold: Firstly, as our previous work [23, 27] was based on models without transverse excitations, which undoubtedly play a key role in the glassy dynamics [5, 13], we want to generalize our field-theoretical treatment to include these degrees of freedom. In the present treatment we assume that they exclusively are affected by the disorder. The general case will be studied elsewhere.

The second aim is to formulate a theory for the energy diffusivity in order to be able to describe the temperature dependence of the thermal conductivity. This is, indeed, possible within the field-theoretical formalism (nonlinear sigma-model [34, 35]), which is the basis of the present approach, by expanding the effective action to quadratic order beyond the SCBA saddle point. If the resulting expression is inserted into the quantum formula for $\kappa(T)$ (including a phenomenological inelastic scattering rate to cut off the Rayleigh divergence), we find an explanation of the plateau of $\kappa(T)$ *as well as the increase beyond the plateau* in terms of the strong scattering by the disorder. The structure in the temperature dependence of κ can therefore be classified to be a harmonic phenomenon, as anticipated by the quoted simulations.

Let us now consider an elastic medium with a mass density m_0 and elastic (Lamé) constants $\lambda = m_0\tilde{\lambda}_0$ and $\mu = m_0\tilde{\mu}$. The transverse elastic constant μ is assumed to vary randomly in space (“transverse elastic disorder”), i.e., $\tilde{\mu} = \tilde{\mu}(\mathbf{r}) = \tilde{\mu}_0 + \Delta\tilde{\mu}(\mathbf{r})$. This leads to a spatial variation of the local longitudinal and transverse sound velocity according to $c_L(\mathbf{r})^2 = \tilde{\lambda}_0 + 2\tilde{\mu}(\mathbf{r})$; $c_T(\mathbf{r})^2 = \tilde{\mu}(\mathbf{r})$. $\tilde{\mu}_0$ is the mean value of $\tilde{\mu}$, and the fluctuation $\Delta\tilde{\mu}(\mathbf{r})$ is assumed to be a random variable with Gaussian statistics of width $\gamma_\mu \propto \overline{(\Delta\tilde{\mu})^2}$ and zero-range spatial correlations. γ_μ has the meaning of the degree of disorder (“disorder parameter”). The frequency-dependent displacement vector obeys an equation of motion $A(z_\xi)\mathbf{u}(z_\xi, \mathbf{r}) = 0$, where the linear operator A is defined by $A(z)_{ij} = -z^2\delta_{ij} - \tilde{\lambda}_0\nabla_i\nabla_j - \nabla_j\tilde{\mu}(\mathbf{r})\nabla_i - \sum_\ell \nabla_\ell\tilde{\mu}(\mathbf{r})\nabla_\ell\delta_{ij}$,

where $z_\xi = \omega + \xi\eta$, with $\xi = \pm 1$ and $\eta \rightarrow 0$.

The problem of calculating the density of states (DOS) $g(\omega)$ and the energy diffusivity $D(\omega)$ from the configurationally averaged Green's functions $\langle G(\mathbf{r}, \mathbf{r}', z_\xi) \rangle = \langle \langle \mathbf{r} | A(z_\xi)^{-1} | \mathbf{r}' \rangle \rangle$ and their products, resp. is analogous to the corresponding problem for electrons in a random potential and can be treated by field-theoretical methods ($\langle \dots \rangle$ denotes the configurational average). The corresponding formalism has been developed in great detail and leads to the nonlinear-sigma-model theory of transport and localization of electrons [35] and classical waves [34].

We perform now the standard steps [27, 34, 35] in order to rederive this theory for transverse elastic disorder. The first is to apply the replica trick [36, 37], which enables one to perform the configurational average from the outset. The resulting effective action contains an interaction with coupling constant γ_μ , which is taken apart by means of a Hubbard-Stratonovich transformation. This introduces effective matrix fields $\Lambda_{i\ell m j}^{\xi\xi'\alpha\alpha'}(\mathbf{r}, \omega)$ (where α are replica labels), which replace the $\Delta\mu(\mathbf{r})$ in the harmonic part of the effective equation of motion. The corresponding operator $A[\Lambda]$, which operates in the Hilbert space made up by the fields $\mathbf{u}^\alpha(\mathbf{r}, z_\xi)$, has matrix elements

$$\begin{aligned} A[\Lambda]_{ij}^{\xi\xi'\alpha\alpha'} &= -z^2 \delta_{ij} \delta_{\xi\xi'} \delta_{\alpha\alpha'} \\ &- \left((\tilde{\lambda}_0 + \tilde{\mu}_0) \nabla_i \nabla_j + \tilde{\mu}_0 \sum_\ell \nabla_\ell^2 \delta_{ij} \right) \delta_{\xi\xi'} \delta_{\alpha\alpha'} \\ &- \sum_{\ell m} \nabla_\ell \left(\Lambda_{i\ell m j}^{\xi\xi'\alpha\alpha'}(\mathbf{r}, \omega) + \Lambda_{i\ell j m}^{\xi\xi'\alpha\alpha'}(\mathbf{r}, \omega) \right) \nabla_m. \end{aligned} \quad (1)$$

As the effective action is now a quadratic form in the $\mathbf{u}^\alpha(\mathbf{r}, z_\xi)$ fields, the latter can be integrated out yielding an effective action that only depends on the matrix fields:

$$S_{eff}[\Lambda] \propto -\frac{1}{2} \text{Tr} \ln A[\Lambda] - \frac{1}{2\gamma_\mu} \text{Tr} \Lambda^2, \quad (2)$$

where the trace runs over all continuous and discrete variables. The saddle-point equations, which are obtained by making S_{eff} stationary, can be solved with \mathbf{r} independent and replica and ξ diagonal matrix fields of the form $\Lambda_{i\ell m j}^{\xi\xi'\alpha\alpha'}(\mathbf{r}, \omega) = -\Sigma(z_\xi) \delta_{\xi\xi'} \delta_{\alpha\alpha'} \delta_{ij} \delta_{\ell m}$, where $\Sigma(z)$ is called self energy and obeys the following self-consistent set of equations:

$$\Sigma(z) = \gamma_\mu \sum_{k < k_D} [\chi_L(k, z) + \chi_T(k, z)]; \quad (3)$$

$$\begin{aligned} \chi_L(k, z) &= k^2 G_L(k, z) = k^2 [-z^2 + k^2 (c_L^2 - 2\Sigma(z))]^{-1}; \\ \chi_T(k, z) &= k^2 G_T(k, z) = k^2 [-z^2 + k^2 (c_T^2 - \Sigma(z))]^{-1}. \end{aligned}$$

The DOS is calculated from the Green's functions G_L and G_T as

$$g(\omega) = \frac{2\omega}{3\pi} \sum_{k < k_D} \text{Im}\{G_L(k, z_+) + 2G_T(k, z_+)\}. \quad (4)$$

Eqs. (3) are a generalization of the self-consistent Born approximation (SCBA) obtained in the "longitudinal"

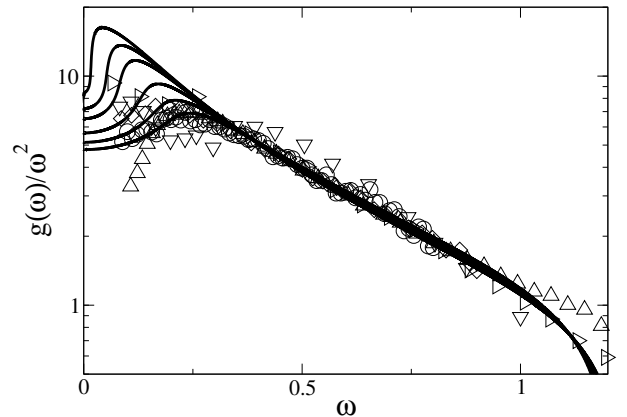


FIG. 1: Lines: Reduced DOS as calculated in SCBA for the disorder parameters (from bottom to top) $\gamma_\mu = 0.13, 0.14, 0.15, 0.16, 0.164, 0.1664$. In all calculations we use units in which $c_T = 1$, $k_D = 1$, $\Theta_D = 1$ (transverse sound velocity, Debye wave number, Debye temperature). The longitudinal sound velocity is set $c_L = \sqrt{2}c_T$. For this choice we have $\gamma_\mu^c = 1/6$. Symbols: measured reduced density of states $g(\omega)/\omega^2$ for some glasses. The frequency has been scaled with a characteristic frequency, and the DOS has been multiplied with a scaling factor to obtain a "universal plot". \blacktriangleright : simulated Lennard-Jones glass [39]; \circ : metallic glass [40]; \triangle : PMMA [41]; \diamond : OTP [42]; ∇ : SiO₂ [43].

version of the present theory [27], and we have $c_L^2 = \tilde{\lambda}_0 + 2\tilde{\mu}_0$ and $c_T^2 = \tilde{\mu}_0$. $\chi_L(k, z)$ and $\chi_T(k, z)$ are the longitudinal and transverse dynamic susceptibility. It is easily seen that Eqs. (3) describes longitudinal and transverse sound-like excitations with dispersions $\Omega_L(q)^2 = [c_L^2 - \text{Re}\{2\Sigma(\omega)\}]q^2$, $\Omega_T(q)^2 = [c_T^2 - \text{Re}\{\Sigma(\omega)\}]q^2$, and a linewidth $\Gamma(q) \propto q^2 \text{Im}\{\Sigma(\omega)\}/\omega$. This is in qualitative agreement with the recent experimental and simulational findings [10, 13]. However the present theory is only valid in the low (ω, q) range, in which the detailed microscopic structure of the material [24, 26, 28] is not important.

As in the models of Refs. [23, 25, 27] the system becomes unstable if γ_μ exceeds a critical value γ_μ^c . For $\gamma_\mu < \gamma_\mu^c$ the reduced DOS $g(\omega)/\omega^2$ exhibits a BP that is enhanced and shifted towards lower frequencies as $\gamma_\mu \rightarrow \gamma_\mu^c$ (see Fig. 1). This critical value ($\gamma_\mu^c = 1/6$ for $c_L^2 = 2c_T^2$) is much smaller than in the longitudinal version of the theory [27] ($\gamma^c = 1/2$), which shows that the transverse excitations are much more sensitive to disorder than the longitudinal ones [38]. As noted previously [23, 27] the BP marks the position where the mean free path (see below) starts to become comparable to the wavelength of the excitations, and where $\Omega(q)$ and $\Gamma(q)$ are of the same order of magnitude.

Why does the BP appear at such a low frequency, although the elastic constants have a symmetric distribution around their mean? As noted already in the literature [16, 23, 25, 27] the disorder affects the vibrational states in such a way that their eigenvalues become distributed like those of a random matrix [36]. The random hybridization of the states leads to level repulsion so that the states "need more space" in the frequency domain than they would without the disorder. So they

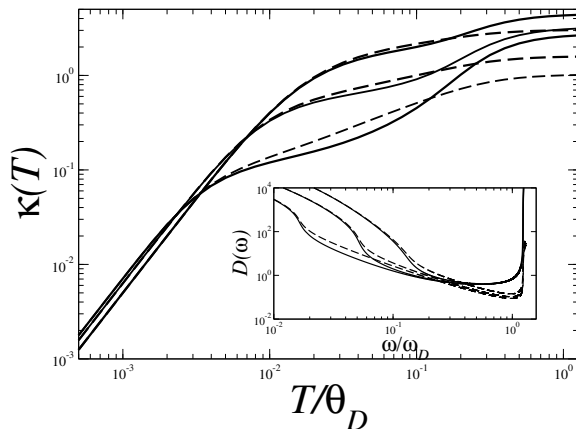


FIG. 2: Insert: Frequency-dependent diffusivity according to the kinetic formula $D(\omega) \propto \ell_0(\omega) \propto 1/[\omega \Sigma''(\omega)]$ (dashed lines) and according to the present theory $D(\omega) \propto \ell_0(\omega)\omega^2/g(\omega)$ (full lines) for the disorder parameters (from right to left) $\gamma_\mu = 0.15, 0.164$, and 0.1664 . Main body: Thermal conductivity $\kappa(T)$ as calculated from Eqs. (3), (5), (8) and (9) using the same $D(\omega)$ as in the insert. The bottom curves correspond to $\gamma_\mu = 0.1664$. For the inelastic scattering parameter we used $C_{in} = 0.02$ in our units. (Taking smaller values of C_{in} results in a shift of the T^2 branch to lower frequencies.)

are both shifted up and down as compared to a Debye spectrum. This effect becomes stronger with increasing γ_μ . The BP marks the low-frequency border of this effect and appears as a shoulder in $g(\omega)$. Only in the $g(\omega)/\omega^2$ representation it appears as a peak.

It has been noted recently [14] that in most of the experimental DOS data $g(\omega)/\omega^2$ exhibits an exponential decrease over a large frequency range. As can be seen from the semilogarithmic representation of Fig. 1 the SCBA is compatible with these findings (see [27]).

Let us now study the impact of the boson peak on the energy diffusivity $D(\omega)$ and the thermal conductivity $\kappa(T)$. The latter can be calculated from the former via

$$\kappa(T) \propto \int d\omega g(\omega) D(\omega) (\omega/T)^2 \frac{e^{\hbar\omega/k_B T}}{[\exp\{\hbar\omega/k_B T\} - 1]^2}. \quad (5)$$

The energy diffusivity $D(\omega)$ can be treated as a classical quantity as long as we do not consider anharmonic interactions. It can be calculated from the Kubo-type formula

$$D(\omega) \propto \lim_{\eta \rightarrow 0} 2\eta \frac{\int d^3\mathbf{r} d^3\mathbf{r}' \langle (x-x')^2 \Delta E(\mathbf{r}, \omega, \eta) \Delta E(\mathbf{r}', \omega, \eta) \rangle}{\int d^3\mathbf{r} d^3\mathbf{r}' \langle \Delta E(\mathbf{r}, \omega, \eta) \Delta E(\mathbf{r}', \omega, \eta) \rangle}, \quad (6)$$

where $\Delta E(\mathbf{r}', \omega, \eta) \propto \sum_{ij} \epsilon_{ij}(\mathbf{r}, z_-)^* \epsilon_{ij}(\mathbf{r}, z_+)$ is the fluctuating part of the energy density with strain tensor $\epsilon_{ij}(\mathbf{r}, z) = \frac{1}{2}[\nabla_i u_j(\mathbf{r}, z) + \nabla_j u_i(\mathbf{r}, z)]$.

In order to be able to evaluate configurational averages of fourfold products of $u_j(\mathbf{r}, z_\xi)$ appearing in Eq. (6) we have to go beyond the saddle-point approximation. Defining the sums $\Lambda_{\alpha\alpha'}^{\xi\xi'} \equiv \sum_{ij} \Lambda_{ij}^{\xi\xi'\alpha\alpha'}$ we put $\Lambda_{\alpha\alpha'}^{\xi\xi'}(\mathbf{r}, \omega) = \Lambda_{sp} + \Delta\Lambda_{\alpha\alpha'}^{\xi\xi'}(\mathbf{r}, \omega)$ and $S_{eff} = S_{eff}^{sp} + \Delta S_{eff}$.

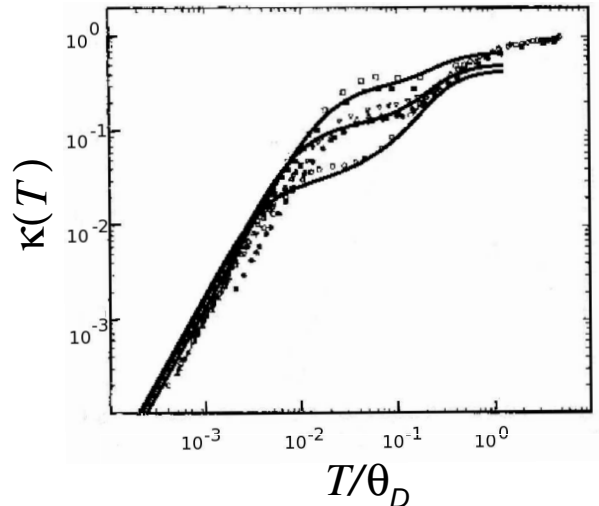


FIG. 3: Scaled thermal conductivity data of several glasses (from top to bottom): PB, PMMA, PET, B₂O₃, PS, SiO₂, as compiled by Freeman and Anderson [6] together with the full-line calculations of Fig. 2.

In terms of the spatial Fourier transforms $\Delta\Lambda_{\alpha\alpha'}^{\xi\xi'}(\mathbf{q}, \omega)$ of $\Delta\Lambda_{\alpha\alpha'}^{\xi\xi'}(\mathbf{r}, \omega)$ this yields a correction to the effective action of the form $\Delta S_{eff} = \sum_{\alpha\alpha'} \sum_{\xi\xi'} \sum_{\mathbf{q}} [\Delta\Lambda_{\alpha\alpha'}^{\xi\xi'}(\mathbf{q})]^* C^{\xi\xi'}(\mathbf{q}, \omega) \Delta\Lambda_{\alpha\alpha'}^{\xi\xi'}(\mathbf{q})$ with $C^{\xi\xi'}(\mathbf{q}, \omega) = \varphi_L^{\xi\xi'}(\mathbf{q}, \omega) + \frac{1}{2}\varphi_T^{\xi\xi'}(\mathbf{q}, \omega) - \frac{1}{2\gamma_\mu}$ and

$\varphi_{L,T}^{\xi\xi'}(\mathbf{q}, \omega) = \sum_{\mathbf{k}} \chi_{L,T}(\mathbf{k} + \frac{\mathbf{q}}{2}, z_\xi) \chi_{L,T}(\mathbf{k} - \frac{\mathbf{q}}{2}, z_{\xi'})$, where the functions $\chi_{L,T}(\mathbf{k}, z_\xi)$ have to be evaluated in SCBA. By inserting a suitable source field into the effective action we can convince ourselves that $C^{+-}(\mathbf{q}, \omega)$ is just the inverse of the Fourier transform of the energy correlation function that appears in Eq. (6) so that we have

$$D(\omega) \propto - \lim_{\eta \rightarrow 0} \eta \frac{1}{C^{+-}(0, \omega)} \left(\frac{\partial}{\partial q_x} \right)^2 C^{+-}(\mathbf{q}, \omega) |_{\mathbf{q}=0}. \quad (7)$$

Evaluating the numerator and denominator explicitly by means of the SCBA expressions, Eqs. (3), of χ_L and χ_T we find

$$D(\omega) \propto \ell_0(\omega)\omega^2/g(\omega) \equiv \tilde{D}(\omega), \quad (8)$$

where $\ell_0(\omega) \propto [\omega \Sigma''(\omega)]^{-1}$ is an effective scattering mean free path and $\Sigma''(\omega) \equiv \text{Im}\{\Sigma(z_+)\}$. Equation (8) is the main result of the present paper. It states that the expression for $D(\omega)$ that corresponds to the SCBA is not equivalent to the kinetic expression $D(\omega) \propto \ell_0(\omega)$ used so far in the literature but contains an “inverse boson peak factor” $\omega^2/g(\omega)$ which reduces the diffusivity in the BP regime in a much stronger way than obtained from the kinetic expression. This is demonstrated in the insert of Fig. 2, where we have compared the frequency dependence of the two formulae. Also the behavior above the BP is quite different in our theory than as obtained from the kinetic formula.

In order to calculate the thermal conductivity by summing over frequency with the proper quantum weight

factor (Eq. (5)), we have to overcome a problem common to all harmonic theories of thermal conductivity, namely the fact that for small frequencies $D(\omega)$ diverges as $D(\omega) \propto \omega^{-4}$ (Rayleigh law). In reality this divergence is cut off by inelastic scattering for which one usually takes the scattering from two-level systems. As we do not deal with anharmonic phenomena in the present paper we use for simplicity

$$\tilde{D}_{eff}(\omega) = [\tilde{D}(\omega)^{-1} + \tilde{D}_{in}(\omega)^{-1}]^{-1} \quad (9)$$

with $\tilde{D}_{in}(\omega)^{-1} = C_{in}\omega$, where C_{in} is a constant which should not vary very much from material to material [7].

In Fig. 2 we compare the results for the thermal conductivity using the kinetic formula and the new theory (Eq. (8)). It is clear that the latter describes the experimentally observed structure much better than the kinetic formula. In Fig. 3 we compare the theory with the experimental results for several glasses as compiled by Freeman and Anderson [6]. It should be mentioned that there is no adjustment or rescaling of the T/θ_D axis. It is clearly seen how the BP is related to the plateau: As the disorder increases the plateau is more pronounced and shifted

to lower temperatures. It is interesting to note that obviously SiO_2 is the material with the strongest disorder and that the Freeman-Anderson scaling may serve as a means to classify glassy materials with respect to their degree of disorder.

In conclusion we have achieved to obtain a mean-field description of the boson peak in terms of transverse elastic degrees of freedom which are affected by the frozen-in disorder. The high-temperature structure of the temperature dependence of the thermal conductivity can be explained by means of this theory as a result of the strong elastic scattering in the frequency regime of the BP and above.

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