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Diffusion of vibrations in disordered systems

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We consider diffusion of vibrations in random lattices with translational invariance. Above some frequency $\omega_{\rm IR}$, corresponding to the Ioffe–Regel crossover (and depending on the strength of disorder), phonons cannot propagate through the lattice and transfer energy. On the other hand, most of the vibrations in this range are not localized. We show that these delocalized excitations are similar to *diffusons* introduced by Allen, Feldman et al. (see, e.g., Phil. Mag. B **79**, 1715 (1999)) to describe heat transport in glasses. In this range the energy in the lattice is transferred by means of diffusion of vibrational excitations. We have calculated the diffusivity of the modes $D(\omega)$ using both the direct numerical solution of Newton equations and the formula of Edwards and Thouless. It is nearly a constant above $\omega_{\rm IR}$ and goes to zero at the localization threshold.

Propagation of vibrational excitations in disordered systems is one of the advanced problems in condensed matter physics. In particular, transport mediated by these excitations is responsible for the thermal conductivity of amorphous dielectrics (glasses). However, mechanisms of heat transfer in glasses above the plateau region are still poorly understood.

At low temperatures below 1 K the low frequency plane long wave acoustical phonons are well defined excitations which transfer the heat in glasses. The thermal conductivity $\varkappa(T) \propto T^2$ and is controlled by a resonant scattering of phonons on the two-level systems (TLS) [1, 2]. Between 4 and 20 K the thermal conductivity $\varkappa(T)$ saturates and displays a well known plateau [3]. As was shown in [4], it can be explained by resonant scattering of phonons by quasilocal vibrations (QLV). The QLV, together with TLS and phonons, are vibrational excitations responsible for many universal properties of glasses [5].

However, above approximately 20 K the thermal conductivity rises again and finally saturates on the level of one order of magnitude higher at the temperatures about several hundreds Kelvin [6]. As generally believed, the origin of this second rise (above the plateau) is not related to phonons. It was established [7], that in this temperature (frequency) range the mean free path of phonons l becomes of the order of their wave length λ (or even smaller, of the order of interatomic distance). Correspondingly, the Ioffe–Regel criterion for phonons [8] is violated. The existence of crossover frequency $\omega_{\rm IR}$ in glasses was predicted in [9] within the framework of the soft potential model and confirmed by molecular dynamics calculations for some real and model glasses [10, 11] and for disordered lattices [12, 13].

In the regime of such strong scattering a standard concept of plane waves (phonons) with well defined wave vector $\mathbf{q}$ becomes inapplicable. If that's the case, what physical mechanism is responsible for the heat transfer in glasses in this temperature range? The numerical simulations show that majority of the vibrational modes in this frequency range are not localized [14–16].

As was shown in [17, 18], a lower limit of the thermal conductivity of amorphous solids above 30 K can be correctly estimated within the framework of the Einstein's model [19]. It was assumed that heat transport above the plateau is a random walk of thermal energy between clusters of neighboring atoms vibrating with random phases. In fact, a diffusion mechanism for the heat transfer in this temperature range was proposed.

At one time, delocalized vibrations in glasses of a new type, different from phonons, were introduced. They were called *diffusons* [20–22]. These are vibrations spreading through the system not ballistically, as phonons, but by means of diffusion. It is an important class of excitations, occupying in glasses the dominant part of the spectrum. The boundary between phonons and diffusons is determined by the Ioffe–Regel crossover frequency $\omega_{\rm IR}$ [22].

In vitreous silica this frequency was estimated to be [10] $\nu_{\rm IR} = 1\,$THz. Integrating density of states [10] up to $\nu_{\rm IR}$, we get for the relative number of phonons $N_{ph} = 0.002 \pm 0.0005$. As a result, in a typical glass only 0.2 % of all vibrational modes are phonons. All other delocalized modes belong to diffusons. Since diffusons are delocalized excitations, they may be responsible for thermal conductivity above the plateau.

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Therefore, it is important to study properties of diffusons systematically in systems where they exist. They bring a new physics to our understanding of vibrational properties in strongly disordered systems and energy/heat transfer in glasses.

Recently, using a random matrix approach, we developed a simple scalar model of a $3d$ disordered harmonic lattice with translational invariance [23, 24]. It show presence of two types of excitations, phonons and presumably diffusons. In the present paper we consider properties of diffusons. To start with, for the sake of clarity of further consideration, we outline below the main properties of our model.

The vibrational properties of a mechanical system of N particles are determined by a dynamical matrix $M_{ij} = \Phi_{ij}/\sqrt{m_i m_j}$, where Φ_{ij} is the force constant matrix and m_i are the particle masses. The matrices M and Φ are real, symmetric, and *positive definite* matrices $N \times N$ (for simplicity we consider a scalar model). The last condition is important. It ensures mechanical stability of the system. For a free mechanical system it is necessary to satisfy also *translation invariance* conditions

$$\sum_i M_{ij} = \sum_j M_{ij} = 0 \tag{1}$$

(for simplicity we consider all masses $m_i = 1$).

We take the dynamical matrix in the form [24]

$$M = AA^{\mathrm{T}} + \mu M_0. \tag{2}$$

Here A is a random matrix $N \times N$ built on a simple cubic lattice with N particles. The only non-zero non-diagonal matrix elements A_{ij} between the nearest neighbors are taken as independent random numbers from a Gaussian distribution with zero mean $\langle A_{ij} \rangle = 0$ and unit variance $\langle A_{ij}^2 \rangle = 1$. The variance controls the degree of disorder. To ensure the translational invariance, the diagonal elements are calculated as follows, $A_{ii} = -\sum_{j \neq i} A_{ji}$. M_0 is a simple crystalline dynamical matrix with unit springs between the nearest neighbors.

If the first term AA^{T} in Eq. (2) is responsible for the disorder in the system, the second term μM_0 describes the ordered part of the Hamiltonian. The parameter μ controls the relative amplitude of this part and the rigidity of the lattice. It can vary in the interval $0 \leq \mu < \infty$. In the paper we have considered the case of strong disorder where $0 \leq \mu \leq 1$ and fluctuating part of the dynamical matrix is bigger than the ordered part. Such form of matrix M guaranties the mechanical stability of the system for any positive value of μ.

The total density of states (DOS) $g(\omega)$, calculated numerically by the kernel polynomial method (KPM) [25] for dynamical matrix (2) for different values of μ, is shown in Fig. 1. As follows from the figure

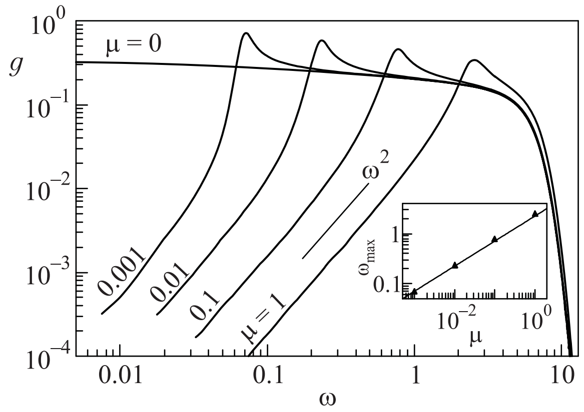

Fig. 1. The normalized to unity DOS $g(\omega)$ for dynamical matrix $M = AA^{\mathrm{T}} + \mu M_0$ and different μ calculated with precise numerical KPM solution for cubic lattice with $N = 200^3$ particles (full lines). The straight line corresponds to ω^2 dependence. Inset: dependence $\omega_{\max}(\mu)$

the DOS, for $\mu = 0$ is nonzero at $\omega \to 0$. In spite of the presence of translational invariance, there are no phonon modes in the lattice with their DOS $g_{ph}(\omega) \propto \omega^2$ for $\omega \to 0$. It means that phonons cannot propagate through the lattice. It is because the rigidity of the lattice and sound velocity for $\mu = 0$ vanish [24].

For $\mu \neq 0$ the DOS at low frequencies is proportional to ω^2. It corresponds to acoustical phonon excitations. Thus, introducing finite values of μ, we open in the spectrum a *phonon gap*. Just above this gap the DOS has a sharp maximum at frequency $\omega_{\max}$ which we will identify with the width of the gap. As follows from the inset, the maximum frequency increases as $\omega_{\max} \propto \sqrt{\mu}$.

As we have shown [26], inside the gap phonons are well defined excitations. Outside the gap they are ill defined, because the Ioffe–Regel crossover frequency $\omega_{\mathrm{IR}} \approx \omega_{\max}$. As we will show in this paper, vibrations outside the gap are delocalized and belong to diffusons.

To determine the localization of the vibrational modes, we have calculated the participation ratio

$$P(\omega) = \left[N \sum_{i=1}^{N} e_i^4(\omega) \right]^{-1}. \tag{3}$$

Here, $e_i(\omega)$ is i-th particle normalized eigenvector with frequency ω. This participation ratio is shown in Fig. 2 for different values of μ. We have checked, that all vibrational modes with exception of small high frequency part are *delocalized*. They have $P(\omega)$ which is independent of the system size. As we show below, for $\mu = 0$ all these modes can be identified as diffusons.

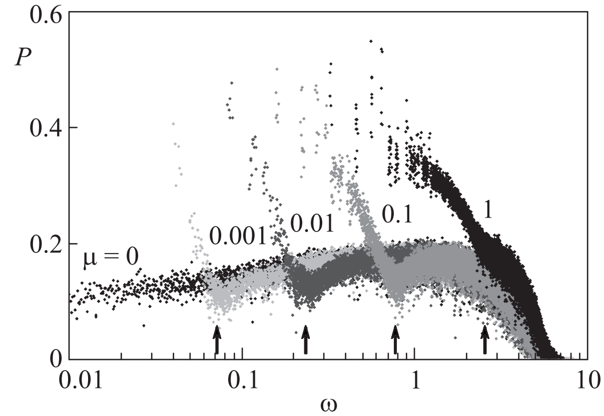

Fig. 2. Participation ratio for different μ as a function of ω for $N = 27^3$ (one realization). The arrows indicate positions of $\omega_{\max}$ in $g(\omega)$ for corresponding values of μ

For $\mu \neq 0$, one can clearly distinguish two different frequency regions in the function $P(\omega)$. As follows from Fig. 1, the low frequency part (below $\omega_{\max}$) corresponds to phonons. In this range the participation ratio increases with decreasing frequency. It is related to increase of the phonon mean free path $l(\omega)$ as $\omega \to 0$. In the high frequency part (above $\omega_{\max}$) $P(\omega)$ is approximately independent of frequency and coincides with $P(\omega)$ for $\mu = 0$. We will show that this range corresponds to diffusons. A similar behavior of the participation ratio was found recently in $2d$ Lennard-Jones glasses [27].

Now let us consider the diffusion of energy in the lattice. The first approach to calculate the diffusivity of energy $D(\omega)$ for vibrations with frequency ω is a direct numerical solution of Newton's equations. For that we have used the Runge–Kutta-4 method with time step $\Delta t = 0.01$ applied to a cubic sample with size L and $N = 10^6$ particles (lattice constant $a_0 = 1$).

Assuming zero initial conditions for displacements and velocities of all the particles, let us apply external forces with frequency ω and random phases φ_i to the particles in the central layer $x = 0$

$$f_i^{\rm ext}(t) = \sin(\omega t + \varphi_i)\exp\left(-t^2/2T^2\right), \qquad (4)$$

where $\omega T \gg 1$. The right and the left sides of the sample have coordinates $x_{r,l} = \pm L/2$. In such a way we excite vibrations with frequencies near frequency ω distributed in a small frequency interval $(\omega - 1/T,\ \omega + 1/T)$. In calculations we used $T = 5$ for all frequencies ω. We started our calculations at time $t_0 = -5T$, when the external force is still negligible.

After applying the force to the central layer, vibrations will spread to the left and to the right ends of the sample. The average squared distance to the energy diffusion front we define as usual

$$R^2(t) = \frac{1}{E_{\rm tot}}\sum_{i=1}^{N} x_i^2 E_i(t). \qquad (5)$$

Here, x_i is the x coordinate of the i-th particle, $E_i(t)$ is the energy of i-th particle and sum is taken over all particles in the sample, $E_{\rm tot} = \sum_i E_i(t)$ is the total energy of the system. It is independent of time after the external force $f_i^{\rm ext}(t)$ becomes negligibly small (i.e., for $t > 5T$).

The energy $E_i(t)$ we define as a sum of the kinetic energy and a half of the potential energy of connecting bonds ($m_i = 1$)

$$E_i(t) = \frac{v_i(t)^2}{2} - \frac{1}{4}\sum_j M_{ij}\left[u_i(t) - u_j(t)\right]^2. \qquad (6)$$

Here, $v_i(t) = \dot{u}_i(t)$ is the particle velocity.

We will apply this method to the case of $\mu = 0$ (i.e., for the lattice without phonons). The results are shown on Fig. 3. As we can see from the figure for small and

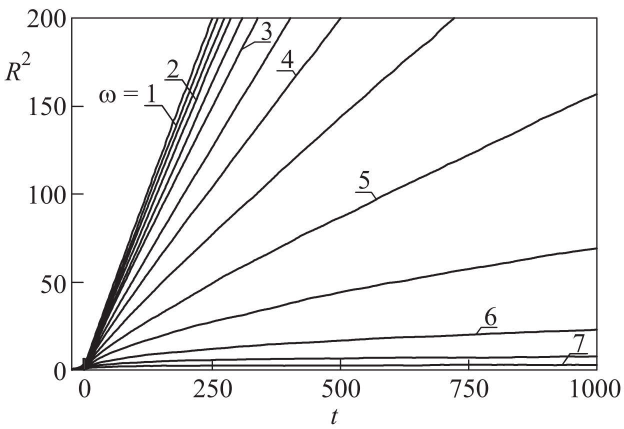

Fig. 3. The dependence of $R^2(t)$ in the case of $\mu = 0$ for one sample with $N = 100 \times 100 \times 100$ particles and 14 different frequencies $\omega = 0.5, 1, 1.5, \ldots, 7$ (from top to bottom). The numbers indicate integer frequencies. The slope of each line corresponds to each black dot in Fig. 4

intermediate frequencies, $R^2(t) \propto t$. Therefore for these frequencies vibrations indeed spread along the x-axis by means of diffusion. The slope of the lines decreases with ω. For calculating the slope, we take time interval Δt when, on the one hand $t > 5T$, and on the other hand, $R \ll L/2$.

From the slope of $R^2(t)$ we can calculate the diffusivity of modes $D(\omega)$ using one dimensional formula

$$R^2(t) = 2D(\omega)t. \qquad (7)$$

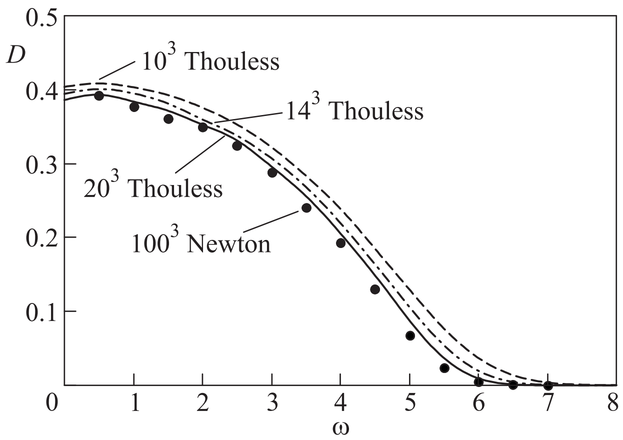

Fig. 4. The diffusivity $D(\omega)$ for $\mu = 0$. Black dots are calculated by the direct solution of Newton's equations from Eqs. (5), (7) and Fig. 3 for $N = 100^3$ particles (one realization). Lines for $N = 10^3, 14^3, 20^3$ are calculated using formula of Edwards and Thouless (8) with $c = 1$ (see below). Averaging for lines is performed over frequencies in the small interval $(\omega - \delta\omega, \omega + \delta\omega)$ with $\delta\omega = 0.25$ and over several thousand realizations

This diffusivity is shown by black dots on Fig. 4. At small frequencies it is approximately a constant, then it decreases with frequency, approaching zero value at the localization threshold, $\omega_{\rm loc} \approx 5.5 \pm 0.5$. At higher frequencies above $\omega_{\rm loc}$ the dependence $R^2(t)$ saturates with increasing t. This indicates to localization of the vibrational modes.

To find the diffusivity $D(\omega)$ for $\mu \neq 0$, the method of numerical solution of Newton's equations is not appropriate. In this case we have phonons in the lattice with long mean free paths. Correspondingly, samples with much bigger sizes are necessary to use this approach. Therefore for $\mu \neq 0$ we used a second approach. The diffusivity $D(\omega_i)$ at eigenfrequency ω_i was calculated by means of the formula of Edwards and Thouless [28], $D(\omega_i) \simeq L^2 \Delta\omega_i$. Here, L is a length of the sample and $\Delta\omega_i$ is sensitivity of ω_i to a twist of boundary conditions. More precisely, we defined the diffusivity as follows:

$$D(\omega) = c \lim_{\varphi \to 0} \frac{L^2}{\varphi^2} \langle |\Delta\omega(\omega)| \rangle, \qquad (8)$$

where φ is angle of the twisting, and c is some constant of the order of unity. It will be determined from comparison with the Newton method. The averaging in Eq. (8) is performed over frequencies ω in the small interval $(\omega - \delta\omega, \omega + \delta\omega)$ with $\delta\omega = 0.25$ and/or over several thousands realizations.

The symmetric real matrix M was defined as usual (2) with periodic boundary conditions. The twisting of the matrix M by angle φ gives a new Hermitian matrix M' obtained as follows. For bonds between the left (l) and the right (r) boundaries of our cubic sample

$$M'_{lr} = M_{lr} \exp(i\varphi), \quad M'_{rl} = M_{rl} \exp(-i\varphi). \qquad (9)$$

For all other bonds $M'_{jk} = M_{jk}$. So $\Delta\omega_i$ is the difference between i-th eigenfrequencies of matrices M and M', $\Delta\omega_i = \omega_i - \omega'_i$. Twisting of boundary conditions was performed for x direction only. For other two directions the periodic boundary conditions were used.

For $\mu = 0$ the results for $D(\omega)$ are shown on Fig. 4 for three different cubic samples (lines). We compared these results with numerical solution of Newton equations for $\mu = 0$ (black dots) and get for the constant $c \approx 1$. Then we used this c value for $\mu \neq 0$. The results are shown on Fig. 5.

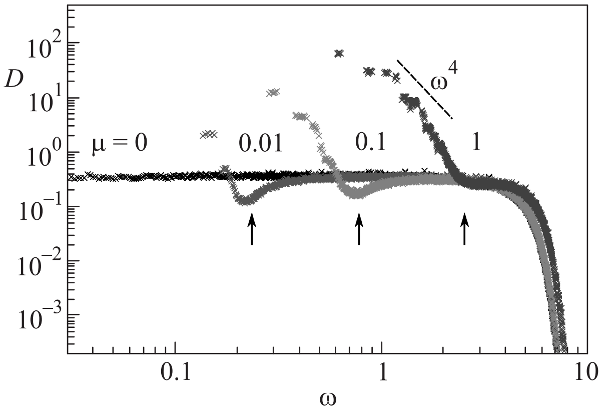

Fig. 5. The diffusivity $D(\omega)$ for various μ (0, 0.01, 0.1, 1) for sample with $N = 14^3$ (crosses). The diffusivity was calculated using formula of Edwards and Thouless (8) with $c = 1$. The arrows indicate frequencies $\omega_{\max}$ in the DOS $g(\omega)$ for corresponding values of μ

For $\mu \neq 0$ we clearly see two different frequency regions in the function $D(\omega)$. At low frequencies, below $\omega_{\max}$, diffusivity increases with decreasing of ω: $D(\omega) = (1/3) l(\omega) v$, v being the sound velocity. This region corresponds to phonons. After a deep minimum at $\omega \approx \omega_{\max}$ the diffusivity $D(\omega)$ saturates on a constant level coinciding with $D(\omega)$ for $\mu = 0$. Therefore $D(\omega)$ in this region corresponds to diffusons. Similar behavior of $D(\omega)$ was found in jammed systems [29, 30]. The deep minimum in the diffusivity at $\omega \approx \omega_{\max}$ corresponds to strong scattering of phonons by quasilocal vibrations near the sharp peak in the DOS (see Fig. 1).

Summarizing, using a random matrix approach we have presented a theory of vibrational properties in strongly disordered systems. In these systems a relative amount of phonons is small and nearly all delocalized vibrations are diffusons. The diffusons play an important role and are responsible for the transport properties of glasses above the plateau. Therefore we think that it is necessary to take them into account in interpretation of experimental data.

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