Vibrations in amorphous solids beyond the Ioffe-Regel criterion

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Abstract. We consider vibrations in 3d random harmonic lattices with translational invariance as a model of amorphous solid. Above some frequency $\omega_{IR}$, corresponding to the Ioffe-Regel crossover, notion of phonons becomes ill defined. They cannot propagate through the lattice and transfer energy. Nevertheless most of the vibrations in this range are not localized. Changing strength of disorder we can vary $\omega_{IR}$ from zero value (when rigidity is zero and there are no phonons in the lattice) up to a typical frequency in the system. Above $\omega_{IR}$ a displacement structure factor $S(q, \omega)$ coincides well with a structure factor of random walk on the lattice. As a result the vibrational line width $\Gamma(q) = D_u q^2$ where $D_u$ is a diffusion coefficient of particle displacements. Our findings may have important consequence for the interpretation of experimental data on inelastic x-ray scattering in glasses.

1. Introduction

Propagation of vibrational excitations in disordered systems is one of the advanced problems in condensed matter physics. In perfect crystals these excitations are well defined plane waves (phonons) having frequency $\omega$ and wave vector $q$. However in amorphous solids the notion of phonons can be lost due to strong scattering on disorder. Then the natural question arises about the mechanism of energy and momentum transfer in this case.

Some time ago, delocalized vibrations in glasses of a new type, different from phonons, were introduced. They were called diffusons [1, 2, 3]. These are vibrations spreading through the system not ballistically, as phonons (on distances of the order of mean free path) but by means of diffusion. It is an important class of excitations which occupy in glasses the dominant part of the spectrum [3]. In these papers it was put forward the hypothesis that the boundary between phonons and diffusons is determined by the Ioffe-Regel criterion for phonons.

Therefore, as we believe, 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. To study these important properties we should have a model, sufficiently simple, to describe all of them.

Recently, using a random matrix approach, we developed a simple scalar model of a 3d disordered harmonic lattice with translational invariance [4, 5] which, as we believe, can represent typical properties of vibrations in disordered systems. It was shown that for certain type of disorder the rigidity of the system is zero and usual phonons cannot propagate through the lattice. However almost all vibrations in the lattice are delocalized and therefore can participate in the heat transfer. To start with, for the sake of clarity of further consideration, we outline below the main properties of our model.
2. A random matrix approach
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 $\Phi_{ij}$ is a force constant matrix and $m_i$ are particle masses. The matrices $M$ and $\Phi$ are real, symmetric and positive definite matrices $N \times N$ (we consider for simplicity 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
\[ \sum_i M_{ij} = \sum_j M_{ij} = 0 \] (for simplicity we consider below all masses $m_i = 1$). It ensures that the potential energy of the system depends only on the differences of particle displacements.

We take the dynamical matrix in the form [5]
\[ M = A A^T + \mu M_0. \] (2)
Here $A$ is a random matrix $N \times N$ built on a simple cubic lattice with $N$ particles and interaction between nearest neighbors only. The only non zero non-diagonal matrix elements $A_{ij}$ between the nearest neighbors are taken as independent random numbers from 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 in the lattice. To ensure the translational invariance the diagonal elements are calculated as a minus sum of non-diagonal elements $A_{ii} = -\sum_{j \neq i} A_{ji}$. $M_0$ 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 $\mu M_0$ describes the ordered part of the Hamiltonian. The parameter $\mu$ controls the relative amplitude of this part and the rigidity of the lattice. It can vary in the interval $0 \leq \mu < \infty$, changing the rigidity and relative amount of disorder. In this paper we have mainly considered the case of strong disorder when $0 \leq \mu \leq 1$ and fluctuating part of the dynamical matrix is bigger then the ordered part. Such form of the dynamical matrix guaranties the mechanical stability of the system for any positive value of $\mu$.

3. Phonons and the Ioffe-Regel criterion
To find the phonon dispersion curve $\omega(q)$ and phonon mean free path $l(\omega)$ we should calculate space and time Fourier transform of the particle displacement field $u(r,t)$. For that we ascribed to all the particles at the initial moment $t = 0$ random displacements (with Gaussian distribution with zero mean and unit variance) and zero velocities. Then, numerically solving Newton equations, we analyzed the particle dynamics at $t \neq 0$.

Let $u(r_i,t)$ be the $i$-th particle displacement as a function of particle coordinate $r_i$ and time $t$. We define the displacement structure factor (DSF) of the displacement field as follows
\[ S(q,\omega) = \frac{2}{NT} \left| \sum_{i=1}^{N} e^{-iqr_i} \int_0^T u(r_i,t)e^{i\omega t} dt \right|^2. \] (3)
For better frequency resolution the upper time limit $T$ was taken sufficiently large. Since vectors $r_i$ in a cubic lattice are discrete, the wave vectors $q \equiv q_n$ are also discrete and are defined on the corresponding reciprocal lattice.

To analyze phonon excitations, we fit the structure factor with the Lorentz distribution
\[ S(q,\omega) \propto \frac{1}{(\omega - \omega_q)^2 + (\Delta \omega)^2}. \] (4)
Figure 1. (a–c) The dependence $\omega_q$ on $q$ for $q \parallel \langle 100 \rangle$ direction for various $\mu$ (1, 0.1, 0.01) in a cubic sample with $N = 50^3$ (one realization). Filled and open diamonds are the maximums of $S(q, \omega)$ as a function of $\omega$ for each discrete value of $q_n$ for frequencies below and above the Ioffe-Regel crossover correspondingly (see text below for details). Solid lines correspond to halves of the maximums. Dashed lines show $\omega = vq$ linear dependence with sound velocity $v$. (d–e) The group velocity $v_g = d\omega/dq$ as a function of $\omega$.

From this fit we can find both the phonon frequency $\omega_q$ and the phonon line width $\Delta \omega$. The results for $\omega_q$ are shown on Fig. 1 for three values of $\mu$ and $q \parallel \langle 100 \rangle$. For sufficiently small values of wave vector $q$ we see a nice linear dispersion curve $\omega_q = vq$.

The phonons are well defined excitations if their mean free path $l = v_g/2\Delta \omega$ exceeds the phonon half wave length $l > \lambda/2 = \pi/q$ (Ioffe-Regel criterion for phonons). We will call the corresponding crossover frequency as $\omega_{IR}$. Fig. 2 shows the ratio $l/\lambda$ as a function of $\omega$ for several values of $\mu$ and different directions of the wave vector $q$. The boundary between filled and open symbols (the full horizontal line) corresponds to frequency $\omega_{IR}$. Thus filled and open symbols on Figs. 1 and 2 belong to phonons with frequencies below and above the Ioffe-Regel crossover frequency correspondingly.

4. Diffusons

Above $\omega_{IR}$ phonons cease to exist as well defined excitations. They are smoothly transformed to diffusons. Since the position of center of inertia is conserved $\sum_i u_i(t) = 0$ (all masses are equal). Therefore we can consider a diffusion of displacements like a diffusion of "particles" in a lattice where the total number of particles is conserved.

Consider first the case of $\mu = 0$ when phonons are absent and only diffusons are present in the lattice. Fig. 3 shows the structure factor $S(q, \omega)$ as a function of wave vector $q$ for three
Figure 2. The ratio \( l(\omega)/\lambda \) as a function of \( \omega \) for different \( \mu \). Different symbols correspond to different \( q \) directions. ◊ for \( q \parallel (100), \triangle \) for \( q \parallel (110), \square \) for \( q \parallel (111) \). The full horizontal line (separating filled and open symbols) corresponds to Ioffe-Regel crossover \( l(\omega) = \lambda/2 \).

Figure 3. The displacement structure factor \( S(q,\omega) \), Eq. (3) (symbols) for \( \mu = 0 \) and for three different frequencies. The sample size is \( N = 50^3 \). The averaging is performed over 300 realizations. Different symbols correspond to different \( q \) directions as explained on Fig. 2. Full lines correspond to the structure factor \( S_{rw}(q,\omega) \) of the random walk on the lattice given by Eq. (5) with \( D_{rw} = 0.7 \). Dashed line corresponds to the limit \( q \ll 1 \) (see Eq. (7)).

different directions in \( q \) space (symbols) and for three different frequencies \( \omega \). Let us compare this displacement structure factor with structure factor of the random walk \( S_{rw}(q,\omega) \) on the lattice. As was shown in [6] for the case of the random walk on a lattice, \( S_{rw}(q,\omega) \) is given by expression

\[
S_{rw}(q,\omega) = \frac{2\Gamma(q)}{\omega^2 + \Gamma^2(q)}, \quad \Gamma(q) = 4D_{rw}\left(\sin^2\frac{q_x}{2} + \sin^2\frac{q_y}{2} + \sin^2\frac{q_z}{2}\right)
\]

(5)

where \( D_{rw} \) is a diffusion constant of the random walk. For small values of \( q \ll 1 \)

\[
\Gamma(q) = D_{rw}q^2
\]

(6)

and in the continuum limit we have the well known result for the diffusion structure factor

\[
S_{rw}(q,\omega) = \frac{2D_{rw}q^2}{D_{rw}^2q^4 + \omega^2}.
\]

(7)
Figure 4. The normalized structure factor $S_n(q, \omega)$ as a function of $q$ for some direction in $q$ space and for each frequency $\omega$ for various values of $\mu$ (0, 0.01, 0.1, 1). The sample size is $N = 50^3$. The averaging is performed over 100 realizations. Left sides of all plots are for $q \parallel (111)$, right sides are for $q \parallel (100)$. White horizontal dashes show the Ioffe-Regel crossover frequency $\omega_{\text{IR}}$. For $\mu = 1$ the frequency $\omega_{\text{IR}}$ is slightly different for different $q$ directions.

A comparison of the displacement structure factor $S(q, \omega)$, (3), and the structure factor of the random walk $S_{\text{rw}}(q, \omega)$, (5), is shown on Fig. 3. One fitting parameter was the diffusion coefficient $D_{\text{rw}}$. From comparison of these data we obtained $D_{\text{rw}} \approx 0.7$. It means that the diffusion coefficient of particle displacements $D_\omega \approx 0.7$. Another fitting parameter was a height $h(\omega)$ of the random walk structure factor in the maximum. According to Eq. (5), in the maximum $\Gamma(q) = \omega$ and $h(\omega) = 1/\omega$, but to fit the data points on Fig. 3 we used slightly higher values of $h(\omega)$. The small difference between $h(\omega)$ and $1/\omega$ can be explained by different frequency dependencies of the density of states for vibrations and for the random walk.

Now let us analyze the displacement structure factor $S(q, \omega)$ for $\mu \neq 0$. For better visual effect we will show a map of the function $S(q, \omega)$ on the plane $(\omega, q)$ for different directions in $q$ space. To do that, for each frequency $\omega$ we have found the maximum $S(q, \omega)$ as a function of $q$ along some directions in $q$ space. Then we normalized function $S(q, \omega)$ along this line $\omega = \text{const}$ to the magnitude of this maximum.

The results are shown on Fig. 4 for four different values of $\mu$ and two directions in $q$ space. The white color corresponds to the maximum when normalized structure factor $S_n(q, \omega) = 1$ while the black color to the case where $S_n(q, \omega) = 0$. For $\mu \neq 0$ we can see clearly two types of excitations in the lattice. At low enough frequencies, below $\omega_{\text{IR}}$, we see phonons with well defined dispersion law $\omega_q$, the same as in the previous Section. At the Ioffe-Regel crossover frequency $\omega_{\text{IR}}$, the structure factor strongly broadens and phonon dispersion line disappears. Above $\omega_{\text{IR}}$ the displacement structure factor coincides well with the structure factor for $\mu = 0$ case shown on Fig. 4a, which corresponds to diffusons. The maximum of the normalized structure factor $S_n(q, \omega)$ (white regions) agrees well with the maximum of the random walk structure factor $S_{\text{rw}}(q, \omega)$ (black line). Deviations from $S_{\text{rw}}(q, \omega)$ take place at high frequencies near the localization threshold.

The vibrational line width above $\omega_{\text{IR}}$ is $\Gamma(q) = D_\omega q^2$ (Eq. 6). Such quadratic dependence of $\Gamma(q)$ was found in many glasses in the experiments on inelastic x-ray scattering, see for example [7, 8] and references therein. It was also found in molecular dynamic simulation of amorphous silicon [9]. However in these and other papers this line width was attributed to phonons without discussion of its physical origin. We guess that the observed $q^2$ dependence of $\Gamma(q)$ has nothing to do with phonons and is in fact related to diffusons.
5. Conclusion
We have found that the delocalized vibrational excitations in this disordered lattice are of two types. At low frequencies below the Ioffe-Regel crossover, $\omega < \omega_{IR}$, they are the usual phonons (plane waves) which can be characterized by frequency $\omega$ and wave vector $\mathbf{q}$ and line width $\Delta \omega$. However, with increasing of $\omega$, due to the disorder-induced scattering, the phonon line width $\Delta \omega$ increases rapidly and at some frequency $\omega \approx \omega_{IR}$ the phonon mean free path $l$ becomes of the order of the wave length $\lambda$.

As a result, at higher frequencies the original notion of phonons is lost and delocalized vibrational modes have a diffusive nature. They are similar to diffusons introduced by Allen and Feldman, et al. [3]. The diffusons again can be characterized by frequency $\omega$, but have no well defined wave vector $\mathbf{q}$. Above $\omega \approx \omega_{IR}$ the structure factor of particle displacements $S(\mathbf{q}, \omega)$ becomes very similar to the structure factor $S_{tw}(\mathbf{q}, \omega)$ of a random walk on the lattice. The former has a broad maximum as a function of $q$ at $q = \sqrt{\omega/D_u}$, where $D_u$ is a diffusion coefficient of the particle displacements.

Presumably diffusons are also accounted for the mysterious $q^2$ dependence of the vibrational line width $\Gamma(q)$ observed in many experiments on inelastic x-ray scattering in glasses. Therefore we think that it is necessary to take them into account in interpretation of experimental data.

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