A random matrix approach to the boson peak and Ioffe-Regel criterion in amorphous solids

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(Dated: July 27, 2020)

We show that the correlated Wishart ensemble can be used to study general vibrational properties of stable amorphous solids with translational invariance. Using the random matrix theory, we found the vibrational density of states and the dynamical structure factor. We demonstrate the presence of the Ioffe-Regel crossover between low-frequency propagating phonons and diffusons at higher frequencies. The reduced vibrational density of states shows the boson peak, which frequency is close to the Ioffe-Regel crossover.

Establishing the general vibrational properties in amorphous dielectrics (glasses) is one of the key problems in the physics of disordered systems. The dominant part of the vibrational spectrum above the Ioffe-Regel crossover and below the mobility edge is occupied by diffusons [1, 2]. These delocalized vibrations are spread by means of diffusive energy transfer from atom to atom. The diffusons are responsible for the heat transfer in glasses in a wide range of temperatures. However, the mechanism of these vibrations is still poorly understood.

Another universal vibrational property of almost all glasses is an excess vibrational density of states (VDOS) well-known as the boson peak. The boson peak was observed using different experimental techniques: the Raman scattering [3, 4], the X-ray scattering [5], the inelastic neutron scattering [6], the far-infrared spectroscopy [7, 8] and the temperature dependence of the heat capacity [10, 13]. Also, the boson peak was observed in two-dimensional structures [11, 15].

There are several theoretical explanations of these anomalies such as an effective medium theory of elasticity [17–21], soft-potential model [22–26], attribution to the transverse-acoustic van Hove singularity [27–29], the mode coupling theory [30]. However, despite a big number of articles about the boson peak, the nature of this phenomenon also remains under discussion [31–33].

It was observed that the boson peak frequency \(\omega_b\) is close to the frequency \(\omega_{ir}\) of the Ioffe-Regel crossover between well-defined phonons with a long mean free path and disordered vibrations, diffusons [34–36]. Therefore, a general theory of the boson peak and Ioffe-Regel criterion can shed light on the nature of vibrations in amorphous solids.

To study these fundamental vibrational features of amorphous solids, we use an approach based on the random matrix theory (RMT). This theory has important applications in many diverse areas of science and engineering [37–40]. Vibrations of amorphous solids are characterized by eigenvalues and eigenvectors of the dynamical matrix \(\mathbf{M}\). The presence of disorder in amorphous systems leads to the random nature of the matrix elements \(M_{ij}\). Therefore, the RMT can be applied to study vibrational properties of amorphous solids [47–50]. It is also applicable to jammed solids [51], which is widely studied nowadays [52, 53]. However, not every random matrix ensemble takes into account special correlations between matrix elements \(M_{ij}\) in amorphous solids. In this work we consider a correlated ensemble, which takes into account only two the most important properties of amorphous solids: (i) the system is near the stable equilibrium position and (ii) the potential energy is invariant under the translation of the system.

In this paper we demonstrate that the general properties (i) and (ii) determine a correlated random-matrix ensemble, which represents the most important properties of amorphous solids like the boson peak and the Ioffe-Regel crossover.

Correlated Wishart ensemble.—The mechanical stability of amorphous solids is equivalent to the positive definiteness of the dynamical matrix \(\mathbf{M}\). Any positive definite matrix \(\mathbf{M}\) can be written as \(\mathbf{M} = \mathbf{A} \mathbf{A}^T\) and vice versa, \(\mathbf{A} \mathbf{A}^T\) is positive definite for any (not necessarily square) matrix \(\mathbf{A}\) [54]. Therefore, we can consider a \(N \times K\) random matrix \(\mathbf{A}\) to obtain a mechanically stable system with the dynamical matrix in the form \(\mathbf{M} = \mathbf{A} \mathbf{A}^T\), which is known as the Wishart ensemble [55, 56]. Each column of the matrix \(\mathbf{A}\) represents a “bond” with a positive potential energy [57]

\[
U_k = \frac{1}{2} \left( \sum_i A_{ik} u_i \right)^2,
\]

(1)

where \(u_i\) is a displacement of \(i\)-th atom from the equilibrium position. Each row of the matrix \(\mathbf{A}\) corresponds to some degree of freedom. The difference between the number of bonds \(K\) and the number of degrees of freedom \(N\) plays a crucial role in vibrational and mechanical properties. In a stable system with a finite rigidity, the number of bonds should be larger than the number of degrees of freedom, which is known as the Maxwell counting rule. For the jammed solids, it was shown that many properties (like the shear modulus and the Ioffe-Regel crossover frequency) are scaled with \(K = (K - N)/N\) [51, 53].

The bond energy \(U_k\) should not depend on the shift \(u_i \rightarrow u_i + \text{const}\). Therefore, the matrix \(\mathbf{A}\) obeys the sum
rule $\sum_j A_{ik} = 0$. It means that the matrix elements $A_{ij}$ are correlated. In the minimal model, we can assume that an amorphous solid consists of statistically equivalent random bonds. In this case the pairwise correlations between matrix elements $A_{ij}$ can be written as

$$\langle A_{ik} A_{jl} \rangle = \frac{1}{N} C_{ij} \delta_{kl}, \quad (2)$$

where $\hat{C}$ is some correlation matrix. One can see that the correlation matrix $\hat{C}$ is proportional to the average dynamical matrix: $\hat{C} = \frac{N}{K} \langle M \rangle$. For simplicity, we consider a scalar model of an amorphous solid on a simple cubic lattice with nearest neighbor interaction. Eigenvalues of the matrix $\hat{C}$ describes a simple cubic lattice with nearest neighbors with a certain rigidity. In this case the matrix $\hat{C}$ has the following structure. The non-diagonal elements $C_{ij} = -\Omega^2$ if atoms with indices $i$ and $j$ are nearest neighbors in the lattice and $C_{ij} = 0$ otherwise. Diagonal elements are $C_{ii} = 6\Omega^2$. The constant $\Omega$ defines a typical frequency in the system.

The correlation matrix $\hat{C}$ is a regular matrix, which describes a simple cubic lattice with nearest neighbor interaction. Eigenvalues of the matrix $\hat{C}$ depend on the wavevector $\mathbf{q}$ which can be expressed as a dispersion law

$$\omega_0^2(\mathbf{q}) = 4\Omega^2 \left( \sin^2 \frac{q_x}{2} + \sin^2 \frac{q_y}{2} + \sin^2 \frac{q_z}{2} \right). \quad (3)$$

Using the random matrix approach, it can be shown that statistical properties of the random matrix $\hat{M}$ are related to the known correlation matrix $\hat{C}$. To find these properties, we consider the corresponding resolvent:

$$\hat{G}(z) = \left\langle \frac{1}{z - \hat{M}} \right\rangle, \quad \hat{G}_0(Z) = \frac{1}{Z - \hat{C}}, \quad (4)$$

where $z$ and $Z$ are complex parameters. The averaging is performed over different realizations of the random matrix $\hat{M}$. In the thermodynamic limit $N \to \infty$ there is a fundamental duality relation between spectral properties of $\hat{M}$ and $\hat{C}$ [58]:

$$Z \hat{G}_0(Z) = z \hat{G}(z), \quad (5)$$

where complex parameters $z$ and $Z$ are related by a conformal map $Z(z)$ defined by the equation

$$zZ + \frac{Z^2}{N} \text{Tr} \hat{G}_0(Z) = z, \quad (6)$$

where parameter $\kappa = (K - N)/N$ defines the relative excess of the number of bonds over the number of degrees of freedom, which controls a proportion between rigidity and disorder in the system. The duality relation makes it possible to find the vibrational density of states (VDOS) and dynamical structure factor (DSF) of the dynamical matrix $\hat{M}$.

**Vibrational density of states.**—To analyze the VDOS $g(\omega)$, we consider the normalized trace of $\hat{G}(z)$, which is the Stieltjes transform of $g(\omega)$:

$$F(z) = \frac{1}{N} \text{Tr} \hat{G}(z) = \int \frac{g(\omega)}{z - \omega^2} d\omega. \quad (7)$$

For regular correlation matrix $\hat{C}$, we can calculate a similar quantity $F_0(Z) = \frac{1}{N} \text{Tr} \hat{G}_0(Z)$. Using the dispersion law for the cubic lattice [3], we find

$$F_0(Z) = \frac{1}{2\Omega^2} W_s \left( \frac{Z}{2\Omega^2} - 3 \right), \quad (8)$$

where $W_s$ is the third Watson integral [59]. On the one hand, from Eq. [5] we know the relation $Z F_0(Z) = z F(z)$. On the other hand, we can express the VDOS as $g(\omega) = \frac{2\omega}{N} \text{Im} F(\omega - i0)$. As a result, we find

$$g(\omega) = \frac{2\omega}{\pi} \text{Im} \frac{1}{Z(\omega^2)}, \quad (9)$$

where the complex parameter $Z$ depends on the real parameter $\omega^2$ through the following complex equation

$$\kappa Z + Z^2 F_0(Z) = \omega^2. \quad (10)$$

This equation defines a contour on a complex plane, which is known as a critical horizon [60]. For a given parameter $\omega$, Eq. [10] has multiple solutions. We choose a physical one with $\text{Im} Z(\omega^2) < 0$ which corresponds to $g(\omega) > 0$.

Equations [8]–[10] defines the VDOS $g(\omega)$ in an implicit form, which can be solved numerically. The result is presented in Fig. 1. For $\kappa = 1$ one can see a low frequency region with the Debye law $g(\omega) \sim \omega^2$. However, for $\kappa = 0$ the VDOS has a constant low-frequency limit. Such behavior of the VDOS was observed in the random matrix model and the jamming transition [41] [53].
The animated plot of the transition between crystalline VDOS \( \kappa = \infty \) and a soft amorphous one \( \kappa = 0 \) is presented in Supplemental Materials [61].

Figure [1] demonstrates a good agreement between the theory and the numerical VDOS calculated for finite interaction radius \( R \) for a system with 400\(^3\) atoms using the Kernel Polynomial Method [62, 63]. The nearest neighbor case \( R = 1 \) was considered before [18, 64]. The generalization of the numerical model for arbitrary \( R \) is presented in Supplemental Materials [61]. As the interaction radius increases, the difference between the theory and the numerical calculation becomes negligible. Therefore, the theory is applicable for a finite interaction radius, which is important to describe amorphous solids.

_Dynamical structure factor._—To analyze the spatial structure of the vibration modes, we calculate the DSF, which specifies the relation between the frequency \( \omega \) and the wavevector \( \mathbf{q} \) [29]. The DSF can be calculated as

\[
S(\mathbf{q}, \omega) = \frac{2\omega}{\pi} \Im \langle \hat{G}(\omega^2) \rangle(\mathbf{q}).
\]  

(11)

Using the duality relation [5] and the dispersion law \( \langle \hat{G}(\omega^2) \rangle(\mathbf{q}) = 1/(\omega^2 - \omega^2_0(\mathbf{q})) \), the resulting dynamical structure factor can be presented in the form of the damped harmonic oscillator (DHO):

\[
S(\mathbf{q}, \omega) = \frac{1}{\pi} \frac{2\omega^2 \Gamma(\mathbf{q}, \omega)}{\omega^2 - \omega^2 E(\mathbf{q}, \omega)^2 + \omega^2 \Gamma^2(\mathbf{q}, \omega)},
\]  

(12)

where the Young modulus is

\[
E(\mathbf{q}, \omega) = \frac{\omega^2_0(\mathbf{q})}{\mathbf{q}^2} \Re \frac{\omega^2}{Z(\omega^2)},
\]  

(13)

and the damping is

\[
\Gamma(\mathbf{q}, \omega) = \frac{\omega^2_0(\mathbf{q})}{\omega^2} \Im \frac{\omega}{Z(\omega^2)} = \frac{\pi}{2} \omega^2_0(\mathbf{q}) g(\omega).
\]  

(14)

Figure [2] shows the normalized DSF for different \( \kappa \). For \( \kappa = 0 \) there is no exact relation between the frequency \( \omega \) and the wavevector \( \mathbf{q} \). Such a broad DSF was attributed to diffusons [2, 49]. For \( \kappa = 1 \) in the low-frequency range there is a linear dispersion \( \omega \sim \mathbf{q} \) with a small broadening due to a small scattering of plane waves. Such low-frequency vibrations are propagating phonons. However, in the dominant frequency range there is a broad behavior of the DSF. Thus, for nonzero \( \kappa \) there is a crossover between phonons and diffusons which is known as the Ioffe-Regel crossover. In this paper we do not consider the Anderson localization which affects only a small part of high-frequency vibrations [2, 49].

_Ioffe-Regel criterion, phonons and diffusons._—To analyze the Ioffe-Regel crossover we consider the low-frequency region \( \omega \ll \Omega \). In this case we can use a small-argument expansion of \( F_0(Z) \). For any three-dimensional system with a linear dispersion \( \omega_0(\mathbf{q}) = \Omega \mathbf{q} \) for \( \mathbf{q} \to 0 \),

\[
F_0(Z) = -a^2 + \sqrt{-Z} \frac{\omega}{4\pi \Omega^3} + O(Z).
\]  

(15)

For the cubic lattice under consideration \( a = \Omega^{-1} \sqrt{\omega_s/2} \), where \( \omega_s \approx 0.505462 \) is a Watson constant [59]. Using Eq. (15), the critical horizon can be found explicitly for \( \omega \ll \Omega \) using an iterative solution of Eq. (10):

\[
\frac{1}{Z(\omega^2)} = \frac{\kappa}{2\omega^2} + \frac{1}{\omega} \sqrt{f(\omega) + \frac{i\omega/4\pi \Omega^3}{\sqrt{\kappa/2 + \omega^2} \sqrt{f(\omega)}}},
\]  

(16)

where \( f(\omega) = \kappa - a^2 \). The sign of \( f(\omega) \) significantly changes the behavior of \( Z(\omega^2) \). The corresponding crossover frequency \( \omega_c = \sqrt{\kappa/2}a \) separates the frequency domain into two regions.

For \( \kappa \ll 1 \) the result [16] can be further simplified for \( \omega < \omega_c \) and \( \omega > \omega_c \), separately. For the VDOS we obtain

\[
g(\omega) = 0 \quad \text{for} \quad \omega < \omega_c,
\]  

(17)

\[
g(\omega) = \frac{2a\sqrt{\omega^2 - \omega_c^2}}{\pi \omega^2 \sqrt{\omega^2 - \omega^2}}, \quad \omega > \omega_c.
\]  

(18)

There is a narrow smooth transition region between [17] and [18]. However, this transition region is much smaller than \( \omega_c \) for \( \kappa \ll 1 \). In the low-frequency region \( \omega \ll \omega_c \), the VDOS has the Debye behavior \( g(\omega) \propto \omega^3 \):

\[
g_D(\omega) = \frac{\omega^2}{2\pi \Omega^3 \kappa \sqrt{\kappa/2}},
\]  

(19)

which corresponds to a static Young modulus \( E_0 = \Omega^2 \kappa \). For \( \kappa = 0 \) the Young modulus becomes zero, which means a soft system without propagation of phonons.
modes with $\omega \ll \omega_c$ the dispersion relation:

$$\omega(q) = \Omega^2 a q \sqrt{2q_c^2 - q^2},$$

where the crossover wavenumber $q_c = \sqrt{\kappa / 2\Omega^2 a^2}$ corresponds to the crossover frequency $\omega_c$. For low-frequency modes with $q \ll q_c$, there is a linear dispersion $\omega(q) = \sqrt{E_0 q}$.

The damping $\Gamma$ follows the vibrational density of states $g(\omega)$ (see Eq. (14)). For $\omega < \omega_c$ it can be written using the dispersion relation:

$$\Gamma = \frac{q^4}{8\pi a} \frac{\sqrt{2q_c^2 - q^2}}{q_c^2 - q^2}. \quad (23)$$

Figure 3 demonstrates the boson peak in the reduced VDOS $g(\omega)/g_D(\omega)$ for different values of the parameter $\kappa$. The boson peak frequency $\omega_b$ is close to the crossover frequency $\omega_c$. For $\kappa \ll 1$ we obtain the relation $\omega_b = \sqrt{3/2}\omega_c$. As a result, the Young modulus $E_0$ is proportional to the boson peak frequency $\omega_b$. This relation was observed by other experimental and theoretical groups [13, 65]. The height of the boson peak is proportional to $\kappa^{-1/2}$, which diverges for $\kappa \to 0$. The boson peak was also observed in two-dimensional systems.

The obtained DSF (12) is defined by the Young modulus $E(q, \omega)$ and the damping $\Gamma(q, \omega)$. For $\kappa \ll 1$ and $q \ll 1$, the Young modulus has a separate form, which depends on the frequency only:

$$E(\omega) = \frac{\Omega^2 \kappa}{2} \left( 1 + \sqrt{1 - \omega^2 \Omega^2 / \omega_c^2} \right), \quad \omega < \omega_c; \quad (20)$$

$$E(\omega) = \frac{\Omega^2 \kappa}{2} + \frac{1}{4\pi \Omega} \left( \frac{\omega}{2\kappa} \right)^{3/2}, \quad \omega > \omega_c. \quad (21)$$

For $\omega < \omega_c$ we can find a dispersion of phonons using the relation $\omega^2/q^2 = E(\omega)$:

$$\omega(\omega) = \Omega^2 a q \sqrt{2q_c^2 - q^2}, \quad (22)$$

where $q_c = \sqrt{\kappa / 2\Omega^2 a^2}$.

The damping $\Gamma$ follows the vibrational density of states $g(\omega)$ (see Eq. (14)). For $\omega < \omega_c$ it can be written using the dispersion relation:

$$\Gamma = \frac{q^4}{8\pi a} \frac{\sqrt{2q_c^2 - q^2}}{q_c^2 - q^2}. \quad (23)$$

For low-frequency modes with $q \ll q_c$, the damping $\Gamma(q) \sim q^4$, which corresponds to the Rayleigh scattering from disorder (Fig. 4). In amorphous bodies, additional resonant scattering of phonons by quasilocal vibrations can occur [26]. However, the number of quasilocal vibrations decreases with increasing relaxation time [67], and this phenomenon goes beyond the general assumptions (i) and (ii) given in the introduction.

The mean free path $l$ is defined by the group velocity $v_g = d\omega(q)/dq$ and damping $\Gamma$ as

$$l = \frac{v_g}{\Gamma} = \frac{16\pi a^2 \Omega^2}{q^4} \frac{(q_c^2 - q^2)^2}{2q_c^4 - q^4}. \quad (24)$$

The mean free path $l$ becomes of the order of the wavelength $\lambda = 2\pi/q$ in the transition region near $\omega_c$ (Fig. 5). It means, that the frequency $\omega_c$ defines the Ioffe-Regel crossover, which is usually written as $l/\lambda \approx 1/2$.

To analyze the DSF in the region $\omega > \omega_c$ we consider the dominant part of this frequency region: $\omega_c \ll \omega \ll \Omega$. In this case, the DSF (12) takes the diffusion form

$$S(q, \omega) = \frac{1}{\pi} \frac{2\Gamma(q, \omega)}{\omega^2 + \Gamma^2(q, \omega)}. \quad (25)$$

which verifies the notion of diffusons introduced in [11, 2]. In the same frequency range $g(\omega) \approx 2\omega/\pi$ and $\Gamma = Dq^2$, where $D = \Omega^2 a$ is a diffusivity. Previously, this form of the DSF was obtained numerically [49].

Figure 4 shows a crossover between the low-frequency Rayleigh scattering $\Gamma \propto q^4$ and the diffusion damping $\Gamma \propto q^2$. Such a quadratic dependence above Ioffe-Regel crossover was observed experimentally [68, 70].

Isostatic state.—If $\kappa = 0$ the number of degrees of freedom $N$ is equal to the number of bonds $K$. In the
jamming transition this state is known as the isostatic state. In this case the macroscopic rigidity becomes zero and the low-frequency VDOS does not follow the Debye law ($\omega_c = 0$). Instead, there is a nonzero low-frequency VDOS. Using the second-order approximation in the random matrix model, we obtain

$$g_{\mathrm{is}}(\omega) \simeq \frac{2a}{\pi} - \frac{1}{4\pi^2U^3} \sqrt{\frac{\omega}{2a^3}}. \quad \text{(26)}$$

This dependence has a linear form as function of $\sqrt{\omega}$ (see inset in Fig. 1). Such a low-frequency cusp-like singularity of the isostatic VDOS was observed numerically in the random matrix model [$\text{51}$] and the jamming transition [$\text{33}$ [$\text{54}$]. In our model, this behavior is related to the diffusion nature of vibrations in this frequency range with the DSF governed by Eq. (25).

In summary, we have demonstrated that the random matrix theory can be applied to study general vibrational properties of amorphous solids. Taking into account only the most important correlations of random matrices, which ensure the mechanical stability (i) and the translation invariance (ii), we find the vibrational density of states and the dynamical structure factor. We demonstrate the presence of the Ioffe-Regel crossover between low-frequency propagating phonons and diffusons at higher frequencies. The boson peak essentially appears near the Ioffe-Regel crossover. The obtained scaling relations correspond to transverse vibrational properties of the jammed solids if we put $\propto \sim z - z_c \sim \Delta \phi^{1/2}$ and $\Omega \sim \Delta \phi^{(\alpha-2)/2}$ [$\text{33}$ [$\text{55}$ [$\text{56}$].

We wish to acknowledge D. A. Parshin, V. I. Kozub, Y. M. Galperin, and V. L. Gurevich for valuable discussions. The work was supported by the Russian Federation President Grant no. MK-3052.2019.2.
