Application of the random matrix theory to vibrational properties of amorphous solids

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Abstract. In this work, we apply the random matrix theory to the study of vibrational properties of disordered systems with a big number of degrees of freedom, such as amorphous solids. Mechanical stability of amorphous solids implies the Wishart ensemble with the positive definite dynamical matrix of the form $M = B B^T$. The translational invariance in amorphous solids leads to the sum rule $\sum_i B_{ij} = 0$, which means non-zero correlations of the matrix elements $B_{ij}$. The proposed correlated Wishart ensemble has many universal properties of amorphous solids. One of these properties is the boson peak in the reduced vibrational density of states $g(\omega)/\omega^2$. The boson peak has been observed in many experiments on amorphous solids, but its nature was not clear so far. We show that the boson peak naturally occurs in the proposed model and find its analytical form.

1. Introduction
The random matrix theory (RMT) has important applications in many diverse areas of science and engineering. The RMT is widely used in the statistical analysis of big data and in the study of complex systems with a big number of degrees of freedom. The RMT was applied to study free probability [1], integrable systems [2], growth models [3], wireless communications [4], signal processing [5], numerical computing [6], economics [7], statistical mechanics [8], and quantum theory [9]. In particular, the RMT was applied to describe complex networks, including random scale-free and small-world networks [10] and the network of synaptic connections between neurons in the brain [11].

In this work, we apply the RMT to study vibrational properties of amorphous systems. One of these universal properties is the boson peak in the reduced density of vibrational states $g(\omega)/\omega^2$ [12]. The boson peak is observed in the Raman scattering [13], X-ray scattering [14], an inelastic neutron scattering [15], and the measurements of the heat capacity $C(T)$ as a maximum in $C(T)/T^3$ [16]. Despite a number of articles about the boson peak, there is still no common physical interpretation.

The boson peak frequency $\omega_b$ is close to the frequency $\omega_{\text{IR}}$ of the Ioffe-Regel crossover from well-defined phonons with a long mean free path to disordered vibrations which spread by means of diffusion (from atom to atom) [17, 18, 19]. The vibrations above the Ioffe-Regel crossover and below the mobility edge was called diffusons [20]. In many amorphous solids, the diffusons occupy the dominant part of the vibrational spectrum. The study of the boson peak can shed light on the nature of vibrations in amorphous solids.
Vibrations of amorphous solids are characterized by eigenvalues and eigenvectors of the dynamical matrix $\hat{M}$. 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 and jammed systems [21, 22, 23, 24]. 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 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.

2. Correlated Wishart ensemble

The mechanical stability of amorphous solids is equivalent to the positive definiteness of the dynamical matrix $\hat{M}$. Any positive definite matrix $\hat{M}$ can be written as $\hat{M} = \hat{B}\hat{B}^T$ and vice versa, $\hat{B}\hat{B}^T$ is positive definite for any (not necessarily square) matrix $\hat{B}$. Therefore, we can consider a $N \times K$ random matrix $\hat{B}$ to obtain a mechanically stable system. Each column of the matrix $\hat{B}$ represents a bond with a positive potential energy

$$\bar{E}_j = \frac{1}{2} \left( \sum_i B_{ij} u_i \right)^2, \quad (1)$$

where $u_i$ is a displacement of $i$-th atom from the equilibrium position. Each row of the matrix $\hat{B}$ 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 Maxwell counting rule. For the jammed solids, it was shown that many properties (like the shear modulus and the crossover frequency) are proportional to $K - N$ [25, 23].

The bond energy $E_j$ should not depend on the shift $u_i \rightarrow u_i + const$. Therefore, the matrix $\hat{B}$ obeys the sum rule $\sum_i B_{ij} = 0$. It means that the matrix elements $B_{ij}$ are correlated. We can assume that the amorphous solid consists of statistically equivalent random bonds. In this case the pairwise correlations between matrix elements $B_{ij}$ can be written as

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

where $\hat{C}$ is some correlation matrix. One can see that the correlation matrix $\hat{C}$ is equal to the average dynamical matrix $\langle \hat{M} \rangle$. For simplicity, we consider an amorphous system as a simple cubic lattice with random bonds. In this case the average dynamical matrix $\langle \hat{M} \rangle$ is a crystalline matrix. It is natural to assume that the crystalline matrix has simple bonds between 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. Otherwise, $C_{ij} = 0$. Diagonal elements are $C_{ii} = 6\Omega^2$. The constant $\Omega$ defines the typical frequency in the system.

3. Vibrational density of states

If we take into account the pairwise correlations only (Eq. (2)), the VDOS can be obtained from the so-called critical horizon [26, 27]. The critical horizon is a contour in the complex plane $Z = X + iY$ defined by the following integral equation:

$$\int \frac{\rho(\varepsilon)\varepsilon^2}{(\bar{X} - \varepsilon)^2 + \bar{Y}^2} d\varepsilon = 1 + \kappa, \quad (3)$$
where $\rho(\varepsilon)$ is the distribution of eigenvalues of the correlation matrix $\hat{C}$ and $\kappa = K/N - 1$. For the case of the simple cubic lattice, we have

$$
\rho(\varepsilon) = \frac{1}{8\pi^3} \int_{-\pi}^{\pi} \int_{-\pi}^{\pi} \int_{-\pi}^{\pi} \delta \left( \varepsilon - 4\Omega^2 \left( \sin^2 \frac{k_x}{2} + \sin^2 \frac{k_y}{2} + \sin^2 \frac{k_z}{2} \right) \right) dk_x dk_y dk_z
$$

$$
= \frac{1}{\pi} \int_0^\infty J_0^2(2\Omega^2 \eta) \cos(\varepsilon \eta - 6\Omega^2 \eta) d\eta, \quad (4)
$$

where $J_0$ is the Bessel function of zero order. The vibrational density of states (VDOS) $g(\omega)$ can be obtained from the critical horizon using the following expressions [26, 27]:

$$
\omega^2 = (X^2 + Y^2) \int \frac{\rho(\varepsilon) \varepsilon}{(X - \varepsilon)^2 + Y^2} d\varepsilon,
$$

$$
g(\omega) = \frac{1}{\pi X^2 + Y^2}. \quad (6)
$$

The analysis of the equations (3)–(6) for $\kappa \ll 1$ shows that the VDOS $g(\omega)$ consists of two regions separated by a crossover frequency $\omega_c$:

$$
\omega_c^2 = \frac{\kappa^2}{4} \left( \int \frac{\rho(\varepsilon) \varepsilon}{\varepsilon} d\varepsilon \right)^{-1} = \frac{\kappa^2}{2W_s} \Omega^2. \quad (7)
$$

In the specific case of the simple cubic lattice (4), $W_s = 96\pi^3 \Gamma^2 \left( \frac{1}{24} \right) \Gamma^2 \left( \frac{11}{24} \right) \approx 0.505462$ is the third Watson integral [28]. Below the crossover frequency ($\omega < \omega_c$), the VDOS has the form

$$
g_\text{b}(\omega) = \frac{\omega \omega_c}{\sqrt{2\pi^2 \Omega^2 \kappa^{3/2}}} \sqrt{1 - \sqrt{1 - \omega^2 / \omega_c^2}}, \quad (8)
$$

In the low-frequencies region $\omega \ll \omega_c$, the VDOS (8) has the Debye behavior:

$$
g_\text{D}(\omega) = \frac{\omega^2}{2\pi^2 \Omega^2 \kappa^{3/2}} + \mathcal{O}(\omega^4). \quad (9)
$$

Above the crossover frequency ($\omega > \omega_c$), the VDOS has the form

$$
g_\text{r}(\omega) = \frac{\kappa}{\pi \omega_c} \sqrt{1 - \frac{\omega_c^2}{\omega^2}}. \quad (10)
$$

Figure 1(a) shows the VDOS for different values of the parameter $\kappa$. One can see that $|g_\text{r}(\omega) - g(\omega)| \ll g(\omega)$ for any frequency $\omega$ except a narrow region near the crossover $\omega_c - \delta < \omega < \omega_c + \delta$ and high-frequency region $\omega \sim \Omega$. As can be seen in Figure 1(b), there is a boson peak in the reduced VDOS $g(\omega)/g_\text{D}(\omega)$ right above the crossover frequency $\omega_c$. Using Eq. (10), we can find the maximum of $g_\text{r}(\omega)/\omega^2$, which gives the boson peak frequency $\omega_b = \sqrt{3/2} \omega_c$.

4. Uncorrelated approximation

We can compare our results with the simplest case of the uncorrelated matrix $\hat{B}$, which does not take into account the sum rule $\sum_i B_{ij} = 0$. In this case we have the classical Wishart ensemble with the trivial correlation matrix $C_{ij} = \Omega_0^2 \delta_{ij}$ with some typical frequency $\Omega_0$. In this case, the VDOS has the Marchenko-Pastur form [29]

$$
g_0(\omega) = \frac{1}{\pi \Omega_0^2 \omega} \sqrt{(\omega_+^2 - \omega^2)(\omega^2 - \omega_-^2)}, \quad \omega_- < \omega < \omega_+, \quad (11)
$$
Figure 1. (a) The VDOS \( g(\omega) \) in the correlated ensemble \( \hat{M} = \hat{B}\hat{B}^T \) for different parameters \( \kappa \). Solid color lines show the numerical solution of Eqs. (3)–(6). Dashed color lines show the Debye VDOS \( g_D(\omega) \). Black solid lines show asymptotic expressions \( g_l(\omega) \) and \( g_r(\omega) \). Vertical dotted lines show the crossover frequency \( \omega_c \). (b) The same for the reduced VDOS \( g(\omega)/g_D(\omega) \).

where
\[
\omega_{\pm} = \Omega_0 \left( \sqrt{1 + \kappa \pm 1} \right). \tag{12}
\]

The VDOS for frequencies smaller \( \omega_- \) and higher \( \omega_+ \) is equal to zero. In the small frequency region we have
\[
g_0(\omega) = \frac{\kappa}{\pi \omega_-} \sqrt{1 - \frac{\omega_-^2}{\omega^2}}. \tag{13}
\]

One can see that \( g_0(\omega) \) is equal to \( g_r(\omega) \) if \( \omega = \omega_- \) and \( \omega \ll \Omega \). Thus, the VDOS above the crossover (\( \omega > \omega_c \)) can be described by the uncorrelated ensemble. Figure 2 shows that the
VDOS $g(\omega)$ coincide well with $g_0(\omega)$ above the crossover frequency except a small difference at high frequencies $\omega \sim \Omega$.

**Figure 2.** The VDOS in the ensemble $\hat{M} = \hat{B}\hat{B}^T$ for different parameters $\kappa$. Color solid lines show the solution of Eqs. (3)–(6) in the correlated case. Color dashed lines show the Marchenko-Pastur law (11) for the uncorrelated case. Vertical dotted lines show the crossover frequency $\omega_c$.

However, the VDOS $g(\omega)$ below the crossover ($\omega < \omega_c$) has a non-trivial behavior $g_l(\omega)$ with the Debye asymptotic (9) while for the uncorrelated ensemble $g_0(\omega) = 0$ for $\omega < \omega_-$. Since the sum rule is the necessary requirement for the existence of plane-wave-like phonons, we can attribute the low-frequency region $\omega < \omega_c$ to phonons. Therefore, the frequency $\omega_c$ can be considered as a crossover frequency between phonons and diffusons. As a result, the frequency $\omega_c$ plays a role of the Ioffe-Regel frequency.

5. Conclusion
Summarizing, we apply the random matrix theory to analyze the correlated Wishart ensemble and find the implicit equation for the vibrational density of states $g(\omega)$. We show, that exist two parts of density of states, $g_l(\omega)$ and $g_r(\omega)$, separated from each other by the crossover frequency $\omega_c$. The left part $g_l(\omega)$ describes the phonon region with Debye law $g(\omega) \sim \omega^2$ at small frequencies. The right part $g_r(\omega)$ describes the diffuson region and corresponds to the Marchenko-Pastur law for the uncorrelated Wishart ensemble. We conclude that crossover frequency $\omega_c$ can be considered as Ioffe-Regel crossover frequency $\omega_{ir}$.

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