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Quasi-local vibrations of amorphous solids in correlated random matrix theory

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Abstract. We apply the random matrix theory to the study of quasi-localized modes in amorphous solids having correlated disorder due to the stability criterion. We demonstrate that the number and properties of quasi-local vibrations depend significantly on how much the statistics of the dynamical matrix elements differs from the Gaussian one. The quasi-localized regime of the vibrational density of states can be understood in the framework of a perturbation theory, which managed to identify the low-frequency asymptotic.

Keywords: amorphous solids, quasi-localized modes, random matrices

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Материалы конференции
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Квазилокальные колебания в аморфных твердых телах в рамках теории коррелированных случайных матриц

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Аннотация. Нами применена теория случайных коррелированных матриц для исследования квазилокальных мод в аморфных твердых телах, имеющих коррелированный беспорядок за счет критерия устойчивости. Мы показали, что количество и свойства квазилокальных колебаний существенно зависят от того, насколько статистика элементов динамической матрицы отличается от гауссовой. Вклад квазилокальных колебаний в плотность колебательных состояний может быть изучен в рамках теории возмущений, которая позволяет найти низкочастотную асимптотику плотности состояний.

Ключевые слова: аморфные тела, квазилокальные колебания, случайные матрицы

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Introduction

Revealing the microscopic nature of the vibrations of strongly disordered mechanical systems is the main goal of past and current research. Examples of such systems are amorphous dielectrics, or glasses. It is well-known that there are two fundamentally different types of delocalized vibrations propagating in glasses: low-frequency plane waves, or phonons, with a certain mean free path and diffusons, which propagate through the system by diffusion energy transfer [1]. The universal properties of amorphous systems critically depend on the transport properties of phonons and diffusons and are evident in many important experimental research methods and practical problems [2, 3].

In addition to phonons and diffusons, the so-called quasi-local vibrations (QLVs) are responsible for the low-frequency properties of amorphous solids at frequencies around 1 THz [4]. These excitations are long-lived vibrations of small groups of atoms, weakly connected with their surroundings [3]. The study of QLVs properties is complicated by the fact that they are difficult to distinguish from the usual Rayleigh scattering of phonons on disorder [5, 6].

As recent studies show, a population of quasi-localized modes is affected by glass cooling processes and its preparation protocol [7–9]. Anomalously soft regions near the loss of stability are inherent in such systems and lead to force constants correlation due to microscopic disorder of glass and mechanical frustration [10, 11]. However, the main features of QLVs are independent of spatial dimension [5, 12], cooling rate [7], microscopic details [5, 13, 14], and even persist in the most deeply supercooled computer glasses [8]. One can assume that there is a universal relation between QLVs and structural correlated disorder in amorphous solids formed during the cooling processes.

There are various theoretical works aimed at investigating QLVs, in particular, the soft-potential model [15–17]. In these works, it was shown that QLVs determine the main contribution to the scattering of acoustic phonons. However, the question about the influence of structural correlated disorder in amorphous solids on QLVs remains open. Since glass-forming systems settle into disordered configurations, their dynamical matrices are naturally characterized by a specific distribution of elements, which so far has received very little attention. In the present paper we shed light on this aspect in the framework of the random matrix theory.

Materials and Methods

In our earlier investigation we applied the random matrix theory approach to detailed study of the low-frequency excitations of strongly disordered systems [18, 19]. In this model the main glasses properties like the boson peak and the Ioffe-Regel crossover between phonons and diffusons are manifested. Now we study how the random matrix theory reproduces the soft regions and how the statistic of matrix element affects the statistic of QLVs.

The vibrations of solids are characterized by the eigenvectors and eigenvalues of the dynamical matrix M, which has a relation with the force constant matrix $M_{ij}=(m_i m_j)^{-1/2}\Phi_{ij}$ [20]. The elements of M have a specific nature depending on the medium in hand. However, the dynamical matrix M has a common structure caused by the symmetrical mechanical properties. One of them is the mechanical stability of the system near its equilibrium position. It leads to the positive definite symmetric M with non-negative eigenvalues which are eigenfrequencies squared ω^2. Second, the potential energy is invariant under the continuous translation of the system which leads to the sum rule $\sum_i M_{ij} = \sum_j M_{ij} = 0$.

Taking into account the above properties, the dynamical matrix M can be written in the form $M = AA^T$. The matrix A plays the role of an incident matrix, whose rows indicate the degrees of freedom and columns indicate the bonds of a system. In general, the matrix A is a rectangular $N\times K$ matrix A. We consider a scalar model in which the displacements of the atoms are collinear and are described by scalar quantities. In this model, the number of atoms is equal to the number of degrees of freedom N. The parameter $\varkappa = (K - N)/N$ defines the relative excess of the total number of bonds K in comparison to the total number of degrees of freedom N. According to the Maxwell counting rule [21], in a stable system with a finite rigidity the number of bonds should be larger than the number of degrees of freedom. Therefore, the parameter $\varkappa$ varies in a wide range $0 \le \varkappa < \infty$ and controls the relationship between stiffness and disorder in the system. For $\varkappa \ll 1$

we obtain a strongly disordered amorphous solid, which is the most important case for the current study. It must be stressed that elements of A correlated by the sum rule $\sum_i A_{ik} = 0$ to satisfy the condition of the translational invariance.

In amorphous solids under consideration, the elements of A have a random nature due to the presence of disorder in such systems. Also, in real amorphous solids short-range interaction between atoms dominates over the far-range interaction, which leads to highly sparse matrix A. In this light, we consider a model of disordered system based on the following rules. For $\varkappa = 0$ the matrix A is the square matrix, build on the simple cubic lattice with the lattice constants $a_0 = 1$ and atomic masses $m_i = 1$. Each atom is placed in a site of lattice, but the interaction between atoms is random, therefore, the elements of A are random numbers. We consider the case of the short-range interaction in which A_{ij} is a nonzero random number only if atoms with indices i and j are nearest neighbors in this lattice. For the simple cubic lattice, the coordination number $n = 6$, therefore, the matrix A has only 6 non-zero non-diagonal elements in every column. The diagonal element $A_{ii} = -\sum_{j\neq i} A_{ji}$.

For $\varkappa \neq 0$ the matrix A is the rectangular matrix. To accurately account the sum rule, we do the following procedure. We take two independent realizations of the square random matrix A constructed in accordance with the above rules, and we add randomly chosen $\varkappa N$ columns of one matrix to the right of the other matrix. This random addition of the new columns corresponds to a random addition of new bonds to the vibrational system. As a result of this addition of bonds, we get a rectangular matrix A with $\varkappa \geq 0$ satisfying the sum rule.

There is a probability that all elements A_{ik} in the ith row of the matrix A have small values. It leads to a soft area, i-atom of which is weakly connected with its surroundings. Such a soft region leads to the formation of the QLV characterized by a small eigenfrequency ω_{qlv}. The density of QLVs is defined by the distribution of ω_{qlv}. We assume that the matrix elements A_{ik} for $k > N$ are zero which has the probability $(1 - \varkappa)^{n+1}$ in the model under consideration.

For small values of matrix elements in the ith row of the matrix A, one can present the matrix A as

$$A = A_0 + V, \tag{1}$$

where the matrix A_0 has all zero elements in the ith row, and the perturbation matrix V has n non-zero elements $v_1, \ldots, v_n$ which correspond to interaction with the nearest neighbor and one element obeying the sum rule $V_{\text{ii}} = -\sum_{j\neq i} V_{ji}$. It must be stressed that matrices save the simple cubic lattice structure for atomic arrangement, and the sum rule is still imposed on them. Taking into account the random matrix model $M = AA^T$, the dynamical matrix takes the next form:

$$M = A_0 A_0^T + A_0 V^T + V A_0^T + V V^T. \tag{2}$$

In the framework of the downfolding method [22], the frequency of QLV ω_{qlv} can be found from (2) to the second-order perturbation theory on the perturbation V:

$$\omega_{\text{qlv}}^2 = \left\{ V H V^T \right\}_{ii}, \tag{3}$$

where

$$H = I - A_0^T \left(A_0 A_0^T \right)^{-1} A_0, \tag{4}$$

and I is $K \times K$ identity matrix. From equation (3) it follows that the frequency of QLV can be written from the sum of quadratic form:

$$\omega_{\text{qlv}}^2 = \sum_{k'l'=1}^{n+1} C_{k'l'} \nu_{k'} \nu_{l'}, \tag{5}$$

where $v_{k'}$ for $k' = 1, \ldots, n+1$ are non-zero elements V_{ik} for $k = 1, \ldots, K$, and the matrix C is the truncation of the matrix H to the set of non-zero elements in the ith row of the matrix V.

The sum rule $V_{ii} = -\sum_{j\neq i} V_{ji}$ corresponds to the rule $v_i = -\sum_{j\neq i} \tilde{v}_j$, where the element v_i is the V_{ii}, and elements $\tilde{v}_j$ are non-zero elements V_{ji} of the ith column of the matrix V. Therefore, equation (5) takes the form:

$$\omega_{\text{qlv}}^2 = \sum_{k'l'=1}^{n+1} \left(C_{k'l'} \nu_{k'} \nu_{l'} - 2 C_{k'i'} \nu_{k'} \tilde{\nu}_{l'} + C_{i'i'} \tilde{\nu}_{k'} \tilde{\nu}_{l'} \right), \tag{6}$$

As a result, we obtain the following low-frequency density of QLVs (QLVDOS):

$$\rho_{qlv}(\omega) = \left\langle \det C^{-1/2} \right\rangle (1-\kappa)^{n+1} \frac{2\pi^{(n+1)/2}\omega^n}{\Gamma\left(\frac{n+1}{2}\right)} \prod_{k'=1}^{n+1} \rho_{k'}(0), \tag{7}$$

where angle brackets denote the averaging over different realizations of the matrix A_0, Γ is the Gamma-function, $\rho_k(v)$ is the probability density function of $v_{k'}$.

Results and Discussion

The theoretical result (7) compared with the vibrational density of states (VDOS) obtained by the full diagonalization of matrices M. We consider the small systems in which the smallest frequency of phonons is limited by the system size. In such systems, the quasi-localized modes are well distinguishable from phonons due to their low number, and the QLVDOS is a low-frequency region $\omega \ll 1$ of VDOS. Also, for additional identification of QLVs, we consider two different cases of statistics of matrix elements A_{ik}: Gaussian and strongly non-Gaussian statistics. This consideration is due to the significant dependence of the number of soft regions formed by small values of matrix elements Aik on its statistics. How it was demonstrated in our early work, for strongly non-Gaussian statistics the number of quasi-localized modes increases in comparison with the Gaussian statistics [19].

Figure 1 shows the numerical calculation of VDOS in the case of Gaussian statistics. We consider the simple case of independent Gaussian random numbers Aik with zero mean and unit variance. In this case, the probability density function of $v_{i'}$ is $\rho_{i'}(0) = 1/\sqrt{2\pi n}$, and $\rho_{k'}(0) = 1/\sqrt{2\pi}$ for $k' \neq i'$. Then, the QLVDOS (7) takes the form

$$\rho_{qlv}(\omega) = \left\langle \det C^{-1/2} \right\rangle (1-\kappa)^{n+1} \frac{2^{(1-n)/2}\omega^n}{\Gamma\left(\frac{n+1}{2}\right)\sqrt{n}}. \tag{8}$$

In Fig. 1, this QLVs contribution $\rho(\omega) \propto \omega^n$ is marked with a solid line. To obtain the coefficient of equation (8) the averaging in $\langle \det C^{-1/2} \rangle$ may be performed numerically. For large enough system size one can show that

$$\left\langle \det C^{-1/2} \right\rangle \propto \kappa^{-(n+1)/2}, \tag{9}$$

since the typical value of the elements of matrix C is proportional to $\varkappa$ if no finite-size effects are present.

In order to set the non-Gaussian statistics of matrix elements and simulate a wide range of different magnitudes of non-zero matrix elements, we multiply the standard Gaussian random number by log-uniform random variable. In this case the random variable has a form

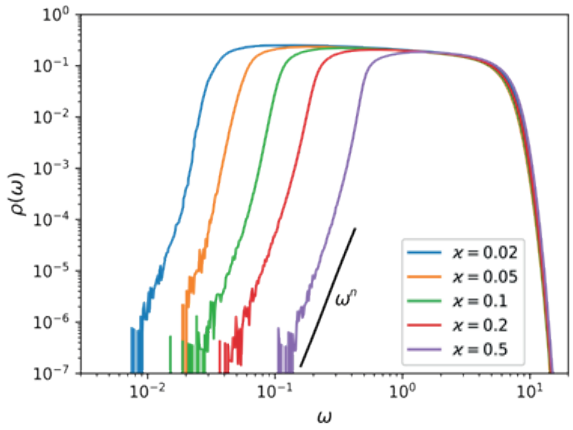

Fig. 1. Vibrational density of states in the case of Gaussian statistics for different parameters $\varkappa$. The solid line marks the QLVs contribution $\rho(\omega) \propto \omega_n$. The matrix A is built on the simple cubic lattice with 10×10×10 atoms

$$\nu_{k'} = c\eta_{k'} \exp \zeta_{k'}, \tag{10}$$

where $\eta_{k'}$ is the Gaussian random number with zero mean and unit variance and $\zeta_{k'}$ is an independent random number uniformly distributed in the interval $(-b/2, b/2)$. The normalization constant $c = \sqrt{(b/\sin h(b)}$ is chosen to provide the unit variance of $v_{k'}$. The resulting probability density function of the random variable $v_{k'}$ has a form

$$p(\nu_{k'}) = \frac{1}{2b\nu_{k'}} \left(erf\left(\frac{\nu_{k'} e^{b/2}}{\sqrt{2c}} \right) - erf\left(\frac{\nu_{k'} e^{-b/2}}{\sqrt{2c}} \right) \right). \tag{11}$$

For $b \gg 1$ it is close to the Gaussian distribution. However, with the increasing of b, the distribution of $v_{k'}$ becomes closer to the reciprocal distribution $\sim 1/|v_{k'}|$, therefore, the parameter b is treated as a non-Gaussianness parameter.

Figure 2 shows an increase in the number of quasi-localized modes with an increase in the non-Gaussian parameter b. As in the case of Gaussian statistics, there is the low-frequency QLVs contribution $\rho(\omega) \propto \omega^n$. The distribution of $v_{k'}$ can be viewed as a mixed distribution of Gaussian random numbers in which the standard deviation is random. Then, the QLVDOS (7) takes the form

$$\rho_{\text{qlv}}(\omega) = \left\langle \det C^{-1/2} \right\rangle (1-\kappa)^{n+1} \frac{2^{(1-n)/2} \omega^n}{\Gamma\left(\frac{n+1}{2}\right)} \left(\frac{2}{b} \sin h \frac{b}{2} \right)^n \left\langle \left(\sum_{k'=1}^{n} e^{2b\zeta_{k'}} \right)^{1/2} \right\rangle, \tag{12}$$

which gives the low-frequency QLVs contribution $\rho(\omega) \propto \omega^n$. Here we use the averaging over different realizations of $\zeta_{k'}$ distributed uniformly in the interval $(-b/2, b/2)$.

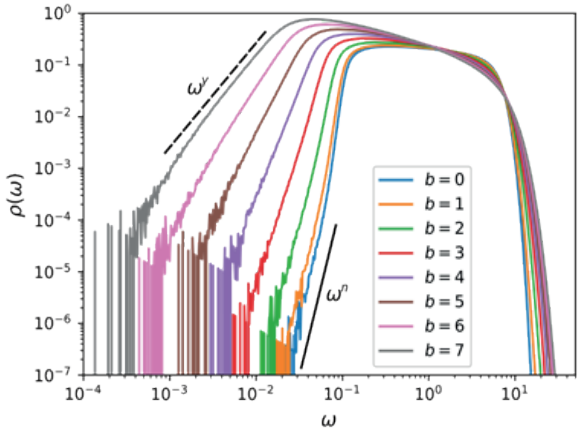

Fig. 2. Vibrational density of states in the case of non-Gaussian statistics with parameter $\varkappa = 0.1$ for different parameters b. The solid line marks the QLVs contribution $\rho(\omega) \propto \omega^n$. The dashed line marks the result $\rho(\omega) \propto \omega^\gamma$ for intermediate range of frequencies. The matrix A is built on the simple cubic lattice with 10×10×10 atoms

However, in contrast to Gaussian statistics, in the case of strongly non-Gaussian statistics the intermediate range of frequencies has additional contribution $\rho(\omega) \propto \omega^\gamma$ with $\gamma < n$. For $b \gg 1$ the random variables v and $\tilde{v}$ are distributed over an exponentially large range of values, thus $\omega \approx \max_k(vk')$ is determined by the maximum value of v, and one can find the next relation for the γ:

$$\gamma = \frac{4n-2}{b} - 1. \tag{13}$$

This expression allows us to describe intermediate degrees of $\rho(\omega)$ for the case of strongly non-Gaussian statistics $b \gg 1$, which corresponds to different slopes in Fig. 2. From the point of view of statistical properties of the matrix A, one can consider the γ as an effective number of neighbors.

Conclusion

Summarizing, we apply the random matrix model to analyze the quasi-local vibrations, which exist in strongly disordered systems like amorphous solids. In this model, the dynamical matrix

is expressed as $M = AA^T$, and numbers and properties of quasi-local vibrations are essentially dependent on the statistic of matrix element A_{ik}. We have shown that as the degree of non-Gaussianness b of the dynamic matrix elements increases, the number of quasi-local vibrations increases too, which manifests itself in their vibrational density of states $\rho_{qlv}(\omega)$. For accurate analysis of quasi-local vibrations, we consider a small system with a low number of phonons, atoms of which are placed in sites of a regular lattice. In a perturbation theory framework we analyzed the low-frequency asymptotic of $\rho(\omega)$ and found that $\rho_{qlv}(\omega) \simeq \omega^n$ for small frequencies with coordination number n, and $\rho_{qlv}(\omega) \simeq \omega^\gamma$ with $\gamma < n$ in the intermediate range of frequencies for strongly non-Gaussian distribution.

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