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Correlation properties of non-affine deformations in the framework of random matrix theory

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Within the framework of the developed model of correlated random matrices, analytical expressions for pair correlations of non-affine components of atomic displacements in a disordered structure are obtained. Correlations of spatial derivatives of non-affine deformations corresponding to correlations between variations in the density of matter arising during deformation are considered. It is shown that such correlations have a delta-shaped component, which corresponds to the statistics of white noise, as well as a negative correlation, exponentially decreasing on the scale of non-affineity.

Keywords: amorphous dielectrics, non-affine deformations, random correlated matrices, Ioffe–Regel length.

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1. Introduction

Studies of the mechanical, vibrational and thermally conductive properties of various amorphous materials with significant disorder at the molecular level have become increasingly important in recent years. Such materials include both glasses and amorphous semiconductors and polymers, as well as nanocomposites based on them. The disordered arrangement of atoms in such materials significantly affects both the behavior of amorphous bodies at nanometer scales and their macroscopic properties. Macroscopic deformations of an amorphous body result in heterogeneous local deformations with characteristic lengthscale of tens of interatomic distances [1,2]. Such inhomogeneous deformations are called *non-affine deformations* since they cannot be described by a combination of local extensions or shears. Non-affine deformations have been observed in many disordered solids: metallic glasses [3], polymeric hydrogels [4], supercooled liquids [5], Lennard–Jones glasess [6], quartz glasses [7]. The origin of non-affine displacements is directly related to structural disorder [4,8], however, this issue requires comprehensive study. In this regard, the description of non-affine deformations and their correlation properties is one of the important tasks of physics of disordered systems, which can shed light on the nature of the amorphous state of matter.

The anomalous mechanical, thermal and acoustic properties observed in amorphous solids are presumably attributable to the presence of spatially inhomogeneous elastic modulus [9]. Due to non-affinity, local bulk and shear moduli exhibit large fluctuations and have distinctive correlation properties [10], which affects the macroscopic elasticity of heterogeneous materials and polymer nanocomposites [11–16]. For example, recent molecular dynamic calculations have directly shown an increase in the local elastic modulus of polystyrene near silica nanoinclusions [12], which is explained by the influence of non-affine deformations.

The amorphous structure becomes metastable due to cooling. This state is characterized by a significant correlation of the force constants of the bonds [17,18]. This results in specific correlations between atomic displacements and inhomogeneous deformations caused by homogeneous mechanical stresses. At the same time, correlations can manifest themselves on large scales, much larger interatomic distances [1,6,19]. The spatial behavior of such correlations can be studied using molecular dynamics calculations, which note the long-range correlation of the non-affine displacement components [1,6,20]. However, the theoretical description of the correlation properties of non-affine deformations is still quite an urgent task.

There are a number of papers devoted to the description of the correlation properties of non-affine deformations. Numerical modeling of a three-dimensional amorphous body was used in Ref. [1] which made it possible to demonstrate the dependence of the correlation of non-affine displacements on distance. The established dependence is decreasing, and there is a range of distances at which correlations have negative values (anticorrelation). The random arrangement of atoms in an amorphous body is taken into account in Ref. [19] for which a function of non-affine displacements and their correlations depending on the distance between the studied points of the system was obtained in the continuum model. However, additional study is needed to identify the relationship between the non-affinity lengthscale and the degree of disorder of the structure.

We applied a model of random correlated matrices in this paper to determine the correlation properties of non-affine deformations. This model made it possible to describe the elastic and vibrational properties of amorphous solids, based on the most general assumptions about the properties of a disordered medium near a stable equilibrium position [11,13,21,22]. The random matrix approach was successfully applied to describe the boson peak and the Ioffe–Regel transition between phonons and the diffusion type of vibrations in systems with different short-range order and different dimensions [21,22]. In particular, the existence of a boson peak in two-dimensional materials was predicted using this approach which was subsequently experimentally confirmed in Ref. [23]. The theory of random matrices made it possible to explain in Ref. [11,13] the appearance of an elastic shell around nanoparticles on the scale of non-affine deformations.

2. Non-affine deformations

Atomic displacements u occur as a response to homogeneous effect when it is applied to the system f, which is described in a linear approximation by the equation of motion [13]:

$$\sum_{j=1}^{N}\left(\Phi_{ij}-\omega^2 m_{ij}\right)u_j = f_i, \tag{1}$$

where $\hat{m}$ — the mass matrix, ω — the vibration frequency of the system, $\hat{\Phi}$ — the force-constants matrix, the elements of which Φ_{ij} are determined by mixed derivatives of potential energy $U(x_1, x_2, \ldots, x_N)$ according to the coordinates of degrees of freedom x_i and x_j:

$$\Phi_{ij} = \frac{\partial^2 U}{\partial x_i \partial x_j}. \tag{2}$$

A scalar approximation is used to simplify the work in which the atomic displacements represent a scalar. This model has already been successfully applied in a number of studies [11,22]. Therefore, the number of degrees of freedom N coincides with the number of atoms of the system, and the indices i and j number the atoms of the system.

The displacement of the i-th atom can be expressed in terms of a resolvent in the following form based on the formula (1)

$$u_i = -\sum_{j=1}^{N} G_{ij} f_j, \tag{3}$$

where $\hat{G}$ — the resolvent of the matrix $\hat{\Phi}$:

$$\hat{G} = \frac{1}{\omega^2\hat{m} - \hat{\Phi}}. \tag{4}$$

The resolvent $\hat{G}$ plays an important role in describing the dynamic properties of amorphous materials. The specific equilibrium coordinates $x_1, x_2, \ldots, x_N$ depend on the cooling process of the melts forming the amorphous material. Therefore, the components of the force-constants matrix and its resolvents depend on the particular system under consideration, and can vary in a broad range [17,18,24]. As a result, the components of atomic displacements also vary, and fluctuations in atomic displacements depend on the strength of disorder in disordered systems.

Let us define the non-affine (fluctuation) component of the displacement u^{na} as the difference between the displacements themselves and their average values:

$$u_i^{\mathrm{na}} = u_i - \langle u_i \rangle, \tag{5}$$

where the brackets $\langle \ldots \rangle$ mean averaging over an ensemble of different realizations of the system in question. The averaged displacement component $\langle u \rangle$ determines the macroscopic response of a substance to external action (macroscopic deformation), and with constant exposure f is expressed in terms of the averaged resolvent:

$$\langle u_i \rangle = -\sum_{j=1}^{N} \langle G_{ij} \rangle f_j. \tag{6}$$

The study of spatial correlation features of non-affine deformations has been of great interest for many years [6,20]. The metastable state in which the amorphous system is located has a significant correlation of force constants, which results in a correlation of non-affine displacement components. This paper considers paired correlations of non-affine components of displacements $\mathscr{R}_{ij}^{\mathrm{na}} = \langle u_i^{\mathrm{na}} u_j^{\mathrm{na}} \rangle$ for studying this issue, which, as follows from equation (5), can be expressed in terms of the resolvent $\hat{G}$ in the following form

$$\mathscr{R}_{ij}^{\mathrm{na}} = \sum_{i',j'=1}^{N} \left(\langle G_{ii'} G_{jj'} \rangle - \langle G_{ii'} \rangle \langle G_{jj'} \rangle\right) f_{i'} f_{j'}. \tag{7}$$

As follows from the entry (7), the presence of non-zero correlations between non-affine deformations is directly related to the existence of paired covariances between the elements of the resolvent $\langle G_{ii'} G_{jj'} \rangle$. In other words, it is necessary to average the resolvent $\langle \hat{G} \rangle$ and study the paired covariances of its elements $\langle G_{ii'} G_{jj'} \rangle$ to study the correlator of non-affine deformations $\mathscr{R}_{ij}^{\mathrm{na}}$. The paper uses a theory based on the use of random correlated matrices for achieving this.

3. Random correlated matrices

The interaction energy U_k, corresponding to the energy of the k-th bond near a stable equilibrium position in the harmonic approximation can be represented as a sum of quadratic forms by atomic displacements:

$$U_k = \frac{1}{2}\left(\sum_{i=1}^{N} A_{ik}u_i\right)^2, \tag{8}$$

where A_{ik} — some numbers, the index k numbers the quadratic forms, which are interpreted as bonds with energy U_k in this paper. Each row of the matrix $\hat{A}$ corresponds to the degree of freedom of the system, and each column corresponds to the relationship between the degrees of freedom. The total potential energy of the interaction

$$U = \sum_{k=1}^{K} U_k, \tag{9}$$

where K is the number of bonds of the system. Then, the force-constants matrix $\hat{\Phi}$ is expressed in terms of the matrix $\hat{A}$ based on equation (2):

$$\hat{\Phi} = \hat{A}\hat{A}^T. \tag{10}$$

The matrix of force constants has a certain form wen the properties of a particular system are described. However, the theory of random matrices is applicable to describe the general universal properties of amorphous systems independent of averaging. As follows from the formula (10), the force-constants matrix is represented in the most general form as an ensemble $\hat{A}\hat{A}^T$ with some matrix $\hat{A}$. At the same time, the matrix $\hat{A}$ has certain properties that follow from the general properties of amorphous disordered systems [24]. The elements A_{ik} are random in nature due to the disorder, and can be considered as random numbers with a given distribution. In the case when the elements of the matrix $\hat{A}$ have a Gaussian distribution, the ensemble $\hat{A}\hat{A}^T$ is called the Wishart ensemble of random matrices [25].

However, it is necessary to consider a correlated Wishart ensemble of random matrices to describe the glassy state, that is, the case when the elements of the matrix $\hat{A}$ are correlated with each other [22]. In general, the paired covariances between the elements of the matrix $\hat{A}$ are given as

$$\langle A_{ik}A_{jl}\rangle = C_{ij}^{kl}, \tag{11}$$

where the matrix of paired covariances $\hat{C}$ has 4 indices: the lower indices i and j number the degrees of freedom of the system, and the upper indices k and l number the bonds. Accounting for correlations results in specific frequency statistics of the amorphous system, which is the subject of many studies [7,22]. The matrix $\hat{A}$ has a sparse appearance due to the short-range interaction between the atoms of the system, characteristic of the amorphous state of matter, which is taken into account in the structure of the correlation matrix $\hat{C}$, which is also sparse. Consideration of the statistical properties of the correlated Wishart ensemble helped to describe the well-known vibration phenomena of amorphous solids, such as the boson peak and the Ioffe–Regel transition during the transformation of phonons into the diffusion type of vibrations [21,22].

As it was shown in Ref. [13] using diagram technique, a system of equations is obtained as a result of averaging over various realizations that links the averaged resolvent $\langle\hat{G}\rangle$ with the covariance matrix $\hat{C}$:

$$\langle\hat{G}\rangle = \frac{1}{\omega^2\hat{I}_N - \hat{\Sigma}}, \quad \Sigma_{ij} = \sum_{k,l=1}^{K} C_{ij}^{kl}\langle G_{kl}^{\star}\rangle,$$

$$\langle\hat{G}^{\star}\rangle = \frac{1}{\hat{I}_K - \hat{\Sigma}^{\star}}, \quad \Sigma_{kl}^{\star} = \sum_{i,j=1}^{N} C_{ij}^{kl}\langle G_{ij}\rangle, \tag{12}$$

where $\hat{G}^{\star}$ — additional resolvent of size $K \times K$, $\hat{\Sigma}$ and $\hat{\Sigma}^{\star}$ — matrices of size $N \times N$ and $K \times K$ respectively, each of which is known in diagram technique as the self–energy part [26], $\hat{I}_N$ and $\hat{I}_K$ — unit matrices of size $N \times N$ and $K \times K$ respectively. Knowing the properties or type of the covariance matrix $\hat{C}$, the properties of the averaged resolvent $\langle\hat{G}\rangle$ can be analyzed from the system of equations (12), which makes it possible to study the general vibrational and mechanical features of the disordered medium [22].

4. Correlation properties of non-affine deformations

The following physical cases are considered in the paper for studying the correlation properties of non-affine deformations, which make it possible to simplify the system of equations (12):

(i) The properties of the disordered system are studied at zero frequency, $\omega = 0$. This further simplifies the system of equations (12). The frequency-dependent properties of non-affine deformations will be the subject of further studies.

(ii) A system is considered in which all its bonds are independent (uncorrelated with each other). This corresponds to the fact that different columns of the matrix $\hat{A}$ representing different bonds have their own correlation matrices:

$$C_{ij}^{kl} = C_{ij}^{k}\delta_{kl}, \tag{13}$$

where δ_{kl} — Kronecker symbol, $\hat{C}^k$ — a matrix describing covariances between degrees of freedom involved in the bonds with the number k. Hereafter, $\hat{C}^k$ denotes a square matrix of size $N \times N$, the elements of which are indexed as C_{ij}^{k}. As follows from the system of equations (12), such consideration results in the diagonal appearance of matrices $\hat{\Sigma}^{\star}$ and $\langle\hat{G}^{\star}\rangle$.

(iii) A homogeneous medium is considered in which all bonds are statistically indistinguishable. In this case, the matrix $\langle \hat{G}^{\star}\rangle$, responsible for describing the bonds of the system, has the following form

$$\langle \hat{G}^{\star}\rangle = \kappa \hat{I}_K, \tag{14}$$

where $\kappa = 1 - N/K$. The relative increase of the number of bonds K over the number of degrees of freedom N is a key parameter in the applied random matrix approach as demonstrated in Ref. [13,21,22], which characterizes the degree of disorder of the system. The parameter $\varkappa = K/N - 1$ was used in these studies. The case $\varkappa \gg 1$ corresponds to weak fluctuations of the matrix $\hat{\Phi}$, and $\varkappa \ll 1$ describes the case of a highly disordered system, the most interesting for consideration.

Taking into account the described properties, using the developed diagram technique described in Ref. [13], we performed averaging over various realizations of the system and obtained an expression for the correlator (7) in the following matrix form:

$$\mathscr{R}^{\rm na}_{ij} = \kappa^2 \sum_{k,l=1}^{K} D^{kl} \sum_{i',j'=1}^{N} L^{kl}_{iji'j'} \langle u_{i'}\rangle \langle u_{j'}\rangle. \tag{15}$$

Here the matrix $\hat{D}$ of size $K \times K$ is given by the expression

$$\hat{D} = \frac{1}{\hat{I}_K - \kappa^2 \hat{T}}, \tag{16}$$

in which the matrix $\hat{T}$ has the size $K \times K$ and is expressed in terms of the averaged resolvent $\langle \hat{G}\rangle$ and the covariance matrix $\hat{C}$:

$$T^{kl} = \mathrm{Tr}\left(\langle \hat{G}\rangle \hat{C}^k \langle \hat{G}\rangle \hat{C}^l\right). \tag{17}$$

The matrix $\hat{L}$ has 6 indices and is also expressed in terms of the averaged resolvent $\langle \hat{G}\rangle$ and the covariance matrix $\hat{C}$:

$$L^{kl}_{iji'j'} = \left(\langle \hat{G}\rangle \hat{C}^k \langle \hat{G}\rangle\right)_{ij} C^l_{i'j'} + \left(\langle \hat{G}\rangle \hat{C}^k\right)_{ii'} \left(\langle \hat{G}\rangle \hat{C}^l\right)_{jj'}. \tag{18}$$

Here and further, the matrices $\langle \hat{G}\rangle$ and $\hat{C}$ are multiplied by the lower indices numbering the degrees of freedom, in accordance with the definition of matrix multiplication, and $\mathrm{Tr}(\ldots)$ means tracing of the matrix by its lower indices. The presence of two terms in the expression (18) is attributable to the fact that two types were identified when analyzing diagrams that make the greatest contribution among all possible diagrams that appear as a result of averaging the paired products of the elements of the resolvent $\langle G_{ii'} G_{jj'}\rangle$.

It is convenient to use the continuous limit to analyze the spatial properties of the found expression for the correlator of non-affine deformations(15). To do this, let us use vector $\mathbf{r}_i$ let's instead of index i that corresponds to the coordinate of the i-th atom. Then the displacements of the atoms $u(\mathbf{r})$ and the studied correlator $\mathscr{R}^{\rm na}(\mathbf{r}, \mathbf{r}')$ are continuous functions depending on the coordinates of the atoms. The action of the matrix $\hat{C}^k$ at the location $\mathbf{r}^k$ of the bond k on a smooth function depending on the coordinates of atoms is characterized by taking the gradient of this function along the coordinate $\mathbf{r}^k$ as shown in Appendix 1 to this paper. Coordinates with upper indices $(\mathbf{r}^k, \mathbf{r}^l)$ denote the coordinates of bonds, and coordinates with lower indices $(\mathbf{r}_i, \mathbf{r}_j)$ denote coordinates of atoms.

Taking into account the replacement of summation over bonds with spatial integration in equation (15), the correlator $\mathscr{R}^{\rm na}(\mathbf{r}_i, \mathbf{r}_j)$ in continuous form has the following form:

$$\mathscr{R}^{\rm na}(\mathbf{r}_i, \mathbf{r}_j) = \frac{\kappa^2 \chi^2}{V^2} \iint D(\mathbf{r}^k, \mathbf{r}^l) L(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}^k, \mathbf{r}^l) d\mathbf{r}^k d\mathbf{r}^l. \tag{19}$$

Here V is the normalization volume, and the constant $\chi = \frac{1}{3K} \sum_{i,j,k} \mathbf{r}_i \cdot \mathbf{r}_j C^k_{ij}$ contains the scalar product of vectors $(\cdot)$. Since $\sum_i C^k_{ij} = \sum_j C^k_{ij} = 0$, the value of χ does not depend on the choice of the origin of the coordinate system for the vectors r_i and r_j and is proportional to the square of the characteristic size of the bond. The function L is a continuous coordinate function and has the following form

$$\begin{aligned} L(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}^k, \mathbf{r}^l) = {} & \nabla_{\mathbf{r}^k} G(\mathbf{r}^k, \mathbf{r}_i) \cdot \nabla_{\mathbf{r}^k} G(\mathbf{r}^k, \mathbf{r}_j) \left(\nabla_{\mathbf{r}^l} \langle u(\mathbf{r}^l)\rangle\right)^2 \\ & + \left(\nabla_{\mathbf{r}^k} G(\mathbf{r}^k, \mathbf{r}_i) \cdot \nabla_{\mathbf{r}^k} \langle u(\mathbf{r}^k)\rangle\right) \left(\nabla_{\mathbf{r}^l} G(\mathbf{r}^l, \mathbf{r}_j) \cdot \nabla_{\mathbf{r}^l} \langle u(\mathbf{r}^l)\rangle\right), \end{aligned} \tag{20}$$

where the operation $\nabla_{\mathbf{r}^k}$ in the Cartesian coordinate system $\{\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z\}$ means taking a gradient along the coordinate $\mathbf{r}^k$:

$$\nabla_{\mathbf{r}^k} = \frac{\partial}{\partial \mathbf{r}^k_x} \mathbf{e}_x + \frac{\partial}{\partial \mathbf{r}^k_y} \mathbf{e}_y + \frac{\partial}{\partial \mathbf{r}^k_z} \mathbf{e}_z. \tag{21}$$

The macroscopic response of the system to external action occurs when deformation is applied to the system. At the same time, in accordance with the macroscopic theory of elasticity, the relative deformation $\boldsymbol{\varepsilon}$, which is a vector in the scalar displacement model, is associated with the gradient of averaged displacements $\langle u\rangle$:

$$\nabla_{\mathbf{r}} \langle u(\mathbf{r})\rangle = \boldsymbol{\varepsilon}, \tag{22}$$

where the relative strain vector $\boldsymbol{\varepsilon} = \{\varepsilon_x, \varepsilon_y, \varepsilon_z\}$ means the stretching of the system along spatial axes. A displacement along the axis z in the direction of which an external deformation will be applied can be considered a displacement in the scalar model. The function L (20) has the following form taking into account that the system is homogeneous and isotropic:

$$\begin{aligned} L(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}^k, \mathbf{r}^l) = {} & \varepsilon^2 \nabla_{\mathbf{r}^k} G(\mathbf{r}^k, \mathbf{r}_i) \cdot \nabla_{\mathbf{r}^k} G(\mathbf{r}^k, \mathbf{r}_j) \\ & + \left(\nabla_{\mathbf{r}^k} G(\mathbf{r}^k, \mathbf{r}_i) \cdot \boldsymbol{\varepsilon}\right) \left(\nabla_{\mathbf{r}^l} G(\mathbf{r}^l, \mathbf{r}_j) \cdot \boldsymbol{\varepsilon}\right). \end{aligned} \tag{23}$$

The form G and D were found as continuous functions in Appendix 2 to this paper. At the same time, it is shown that these functions are differential, that is, depend on the difference of spatial coordinates:

$$G(|\mathbf{r}^k - \mathbf{r}_i|) = -\frac{1}{4\pi\kappa\chi|\mathbf{r}^k - \mathbf{r}_i|}, \tag{24}$$

$$D(|\mathbf{r}^k - \mathbf{r}^l|) = \frac{e^{-|\mathbf{r}^k - \mathbf{r}^l|/\xi}}{4\pi\kappa\xi^2|\mathbf{r}^k - \mathbf{r}^l|}. \tag{25}$$

The parameter ξ defines a certain spatial scale and is set by the expression

$$\xi = \sqrt{\frac{\kappa}{6K}\sum_{m,l} T^{ml}(\mathbf{r}^m - \mathbf{r}^l)^2}. \tag{26}$$

The expression (26) coincides with the parameter of the heterogeneity scale of an amorphous medium obtained in the work [13]. At the same time, as it was shown in Appendix 2, for $\kappa \ll 1$, the scale ratio $\xi \propto \kappa^{-1/2}$ is met, which is characteristic of the non-affinity scale [13] and of the Ioffe–Regel length lengthscale l_{IR}, which is the free path length of phonons near the Ioffe–Regel frequency [22]. In other words, the parameter ξ (26) corresponds to the scale of heterogeneity (non-affinity) of an amorphous system and is associated with the strength of disorder.

Taking into account the found ratios (20)–(26), the displacement correlator (19) has the following form:

$$\mathscr{R}^{\mathrm{na}}(|\mathbf{r} - \mathbf{r}|) = -\frac{\varepsilon^2}{4\pi\kappa|\mathbf{r} - \mathbf{r}|} + \iint \frac{\left((\mathbf{r}^k - \mathbf{r}_i)\cdot\boldsymbol{\varepsilon}\right)\left((\mathbf{r}^l - \mathbf{r}_j)\cdot\boldsymbol{\varepsilon}\right) e^{-|\mathbf{r}^k - \mathbf{r}^l|/\xi}}{(4\pi)^3\kappa V^2\xi^2|\mathbf{r}^k - \mathbf{r}_i|^3|\mathbf{r}^l - \mathbf{r}_j|^3|\mathbf{r}^k - \mathbf{r}^l|}\, d\mathbf{r}^k d\mathbf{r}^l. \tag{27}$$

The first term in the expression (27) gives the main contribution to the considered correlations and does not contain characteristic spatial scales. This result is consistent with the results provided in Ref. [19], according to which the correlation of non-affine deformations $\langle u^{\mathrm{na}}(\mathbf{r})u^{\mathrm{na}}(\mathbf{r}')\rangle$ at points located at a distance $r = |\mathbf{r} - \mathbf{r}'|$ from each other has a spatial dependence $\langle u^{\mathrm{na}}(r)u^{\mathrm{na}}(0)\rangle \propto C - Dr^{-1}$, where C and D — positive constants. At the same time, the first term does not contain a characteristic scale of ξ and cannot fully describe the correlation properties of non-affine deformations of an amorphous body. The characteristic heterogeneity lengthscale of the medium ξ in expression (27) is a part of the second term which was obtained for the first time.

The found correlator of non-affine deformations (27) has a long-range character, which is attributable to the fact that the local deformation of matter is determined not only by the field of non-affine displacements $u^{\mathrm{na}}(\mathbf{r})$ itself, but also by its spatial derivatives. We considered correlations of gradients of non-affine displacements $\langle\nabla_{\mathbf{r}_i}u^{\mathrm{na}}(\mathbf{r}_i)\cdot\nabla_{\mathbf{r}_j}u^{\mathrm{na}}(\mathbf{r}_j)\rangle$ for the scalar displacement model used. Consideration of such correlations simplifies the found expression (27) for $\mathscr{R}^{\mathrm{na}}$:

$$\nabla_{\mathbf{r}_i}\cdot\nabla_{\mathbf{r}_j}\mathscr{R}^{\mathrm{na}}(|\mathbf{r}_i - \mathbf{r}_j|) = \frac{\varepsilon^2}{\kappa}\,\delta(\mathbf{r}_i - \mathbf{r}_j) - \frac{\varepsilon^2}{4\pi\kappa\xi^2}\frac{e^{-|\mathbf{r}_i - \mathbf{r}_j|/\xi}}{|\mathbf{r}_i - \mathbf{r}_j|}. \tag{28}$$

The first term is different from zero only if $\mathbf{r}_i$ and $\mathbf{r}_j$ (delta-correlated component) coincide as can be seen from equation (28). The second term decreases exponentially with the distance $r = |\mathbf{r}_i - \mathbf{r}_j|$ on the non-affinity lengthscale ξ, that is, it expresses a long-range correlation.

5. Discussion of results

Elastic properties of amorphous solids substantially depend on microscopic non-affine deformations. The spatial correlations of non-affine deformations $\langle u^{\mathrm{na}}(\mathbf{r})u^{\mathrm{na}}(\mathbf{r}')\rangle = \mathscr{R}^{\mathrm{na}}(|\mathbf{r} - \mathbf{r}'|)$ are studied using the random matrix model and their analytical expressions are obtained. An important result of the study is the found expression (28) for correlations between spatial derivatives of non-affine deformations, which from a physical point of view can be compared to correlations between local variations in the density of the substance of the system. In general, it was found that the correlations $\langle\nabla u^{\mathrm{na}}(\mathbf{r})\cdot\nabla u^{\mathrm{na}}(\mathbf{r}')\rangle$ rapidly decrease in space and depend on the distance $r = |\mathbf{r} - \mathbf{r}'|$ in the following form

$$\langle\nabla u^{\mathrm{na}}(r)\cdot\nabla u^{\mathrm{na}}(0)\rangle = A\delta(r) - B\,\frac{e^{-r/\xi}}{r}. \tag{29}$$

The resulting correlator consists of a delta-correlated component corresponding to white noise statistics and an exponentially decreasing contribution, expressing long-range correlation decreasing on the non-affinity lengthscale ξ. Such exponential behavior of correlations on the heterogeneity lengthscale ξ was not been observed in the studies of other scientific groups known to us and was obtained for the first time in this work.

The paper shows that the developed theory of random matrices is a good theoretical tool for studying various universal features of amorphous systems with varying degrees of disorder at the molecular level. Previously, the random matrix approach was applied to the theoretical description of the boson peak and the Ioffe–Regel transition between phonons and the diffusion type of vibrations in systems of a random topology and dimension [21,22,24]. The theoretical results obtained helped to substantiate the experimental dependences previously found, including in the study of the boson peak in two-dimensional systems [23,27]. At the same time, it was found that the frequencies of the Ioffe–Regel crossover and boson peak have similar values, and the corresponding spatial scale l_{IR}, usually amounting to several nanometers, is associated with the strength of disorder in such systems and depends on the system parameter $l_{\mathrm{IR}} \propto \kappa^{-1/2}$. It is this lengthscale that determines the magnitude of non-affine deformations in amorphous systems. The scale ratio $\xi \propto \kappa^{-1/2}$ obtained in this study suggests that ξ corresponds to the heterogeneity lengthscale of the medium under study (the non-affinity lengthscale). The classical (continuum) theory of elasticity becomes inapplicable at such scales, since it is impossible to determine a smooth dependence of a displacement on the coordinate. In other words, the non-affinity lengthscale ξ and the Ioffe–Regel length l_{IR} have the same order of magnitude and separate macroscopic scales, to which the classical (continuum) theory of elasticity is applicable, and microscopic scales, on which the disorder of the system plays an essential role, which is consistent with the results of the paper [11,13,22]:

$$\xi \sim l_{\mathrm{IR}} \propto \kappa^{-1/2}. \tag{30}$$

The results of this study are of great importance for the physics of nanocomposites. It was previously shown that in a highly disordered medium with rigid inclusions of nanometer sizes, an effective rigid shell is formed around such nanoclusions, for which the values of elastic modules exceed the values of volumetric elastic modules and depend on the degree of disorder [12,13]. For example, calculations using the method of molecular dynamics of polystyrene with a nanoparticle SiO_2 show an increase of stiffness at a distance of about 1.4 nm around the nanoparticle [12]. At the same time, the scale that determines the thickness of such a shell exactly corresponds to the non-affinity lengthscale ξ obtained in this study. This results in the increase of the effective volume of nanoparticles and in the increase of their effect on the macroscopic elastic properties of nanocomposites, which is especially noticeable with similar values of the linear sizes of nanoparticles and the heterogeneity scale ξ. At the same time, an exponential decrease of elastic modules near a rigid nanoparticle is observed, depending on the distance to it. This fact highlights the relationship between local elastic properties and non-affine deformations in systems with strong disorder. A similar behavior of elastic modules is observed near the transition between amorphous and crystalline layered structures [11]. The presence of transitional phases between structurally different regions is a manifestation of the heterogeneity of local deformations and is directly related to the disorder in such structures.

The original results obtained in this study of spatial correlations of non-affine deformations can be confirmed by molecular dynamic calculations. Molecular dynamic modeling allows considering various amorphous and polymer systems, each of which can be characterized by its ownnon-affinity lengthscale ξ. The non-affinity radius for other amorphous substances was estimated to be about ten typical interatomic or intermolecular distances [1,7]. The dependence of correlations of non-affine deformations can be compared with the theoretical dependence for the obtained molecular dynamics data (29). However, it is worth taking into account the vector nature of the displacements and considering the divergence and rotor correlators of the field of non-affine displacements. The former of them corresponds to the correlation between variations in the density of matter arising during deformation, and the latter corresponds to the correlation between local rotations of matter. It is expected that such correlators will qualitatively have the found dependence (29) for the scalar displacement model. At the same time, A, B and ξ are fitting parameters. It is possible to estimate the heterogeneity lengthscale of the structure ξ from the comparison of the results. We are actively working in this field, and the results of this work will be reviewed by us in the near future.

The relations obtained within the framework of the theory of random matrices can help to study the correlation properties of non-affine deformations at a nonzero frequency $\omega \neq 0$. This is especially relevant in the study of viscoelastic vibrational properties of amorphous systems. It is also of interest to study the correlation properties of non-affine deformations near the boundaries between media with different values of volumetric elastic modulus, which is especially important for nanocomposite amorphous systems. At the same time, each k-th bond of the system should be characterized by its own parameter γ_k for a correct description of the features of such systems in accordance with the paper [13]. The obtained ratios can also help in solving this problem.

6. Conclusion

The correlation properties of non-affine deformations in amorphous solids were studied in this paper. Analytical expressions were obtained using the random matrix model and the developed diagrammatic technique for the paired correlator of non-affine displacements in the approximation of zero frequencies and a homogeneous isotropic disordered system far from its boundaries. The obtained matrix relations were analyzed in the continuous limit, where each atom or bond is characterized by its positions in space, and the functions acting on them are smooth and continuous.

It was shown that the correlator of non-affine displacement gradients consists of a delta-correlated component (white noise) and an exponentially decreasing long-range correlations. The characteristic scale ξ, standing in the exponent, describes the heterogeneity lengthscale of the medium under study and is associated with the disorder parameter κ by the lengthscale ratio $\xi \propto \kappa^{-1/2}$. The results obtained play an important role in studying the viscoelastic properties of amorphous bodies.

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Conflict of interest

The authors declare that they have no conflict of interest.

Appendix 1

Let us consider the action of the matrix $\hat{C}^k$ at the location $\mathbf{r}^k$ of the bond k on some smooth function $h(\mathbf{r}_i)$, which depends on the coordinate of the i-th atom $\mathbf{r}_i$. Let us decompose $h(\mathbf{r}_i)$ near the point k for this purpose:

$$h(\mathbf{r}_i) \simeq h(\mathbf{r}^k) + \sum_{\alpha=\{x,y,z\}} \frac{\partial h(\mathbf{r}^k)}{\partial \alpha} (\mathbf{r}_i - \mathbf{r}^k)_\alpha, \tag{A.31}$$

where the index α numbers the spatial Cartesian coordinates and denotes the projection of the vector.

Let us take into account an important property of translational invariance, which imposes a restriction on the form of the force-constants matrix in the form of the sum rule $\sum_i \Phi_{ij} = \sum_j \Phi_{ij} = 0$. This rule follows from the fact that the potential energy of the system associated with the $\hat{\Phi}$ ratio (2) does not change when the system is shifted as a whole, i.e. it is invariant when u_i is replaced by $u_i + \text{const}$. This leads to the fact that the rule of sum is superimposed on the covariance matrix as follows from the formulas (10)−(11):

$$\sum_{i=1}^{N} C^k_{ij} = \sum_{j=1}^{N} C^k_{ij} = 0. \qquad \text{(A.32)}$$

Then, taking into account the ratios (A.31)−(A.32), the action of the matrix $\hat{C}^{(k)}$ at the location $\mathbf{r}^k$ of the bond k on $h(\mathbf{r}_i)$ can be written as

$$\sum_i C^k_{ij} h(\mathbf{r}_i) = \sum_\alpha \frac{\partial h(\mathbf{r}^k)}{\partial \alpha} \sum_i r_{i\alpha} C^k_{ij}. \qquad \text{(A.33)}$$

Similarly, considering two smooth coordinate functions $h_1(\mathbf{r}_i)$ and $h_2(\mathbf{r}_j)$, the following relation can be obtained:

$$\sum_{i,j} C^k_{ij} h_1(\mathbf{r}_i) h_2(\mathbf{r}_j) = \sum_{\alpha,\beta} \frac{\partial h_1(\mathbf{r}^k)}{\partial \alpha} \frac{\partial h_2(\mathbf{r}^k)}{\partial \beta} \chi^k_{\alpha\beta}. \qquad \text{(A.34)}$$

in which

$$\chi^k_{\alpha\beta} = \sum_{i,j} r_{i\alpha} r_{j\beta} C^k_{ij} \qquad \text{(A.35)}$$

it is a tensor of the second rank. In the case of an isotropic medium, the tensor $\chi^k_{\alpha\beta}$ is diagonal with equal components. In addition, when considering a homogeneous medium, when the bonds do not differ from each other, the tensor $\chi^k_{\alpha\beta}$ does not depend on the number of bond k:

$$\chi^k_{\alpha\beta} = \chi \delta_{\alpha\beta}, \quad \chi = \frac{1}{3K} \sum_{ijk} \mathbf{r}_i \cdot \mathbf{r}_j C^k_{ij}, \qquad \text{(A.36)}$$

where $(\cdot)$ denotes the scalar product of vectors. Thus, taking into account the expression (A.36), the equation (A.34) can be presented in the following form:

$$\sum_{i,j} C^k_{ij} h_1(\mathbf{r}_i) h_2(\mathbf{r}_j) = \chi \nabla_{\mathbf{r}^k} h_1(\mathbf{r}^k) \cdot \nabla_{\mathbf{r}^k} h_2(\mathbf{r}^k), \qquad \text{(A.37)}$$

where the operation $\nabla_{\mathbf{r}^k}$ in the Cartesian coordinate system $\{\mathbf{e}_x,\ \mathbf{e}_y,\ \mathbf{e}_z\}$ means taking a gradient along the coordinate $\mathbf{r}^k$:

$$\nabla_{\mathbf{r}^k} = \frac{\partial}{\partial r^k_x} \mathbf{e}_x + \frac{\partial}{\partial r^k_y} \mathbf{e}_y + \frac{\partial}{\partial r^k_z} \mathbf{e}_z. \qquad \text{(A.38)}$$

The found ratio (A.37) also occurs when considering the action of the matrix $\hat{C}^k$ at the location $\mathbf{r}^k$ of the bond k on a smooth spatial function $h(\mathbf{r}_i, \mathbf{r}_j)$, which depends on two coordinates of atoms i and j:

$$\sum_{ij} C^k_{ij} h(\mathbf{r}_i, \mathbf{r}_j) = \chi \nabla_{\mathbf{r}_i} \cdot \nabla_{\mathbf{r}_j} h(\mathbf{r}_i, \mathbf{r}_j)\Big|_{\mathbf{r}_i=\mathbf{r}_j=\mathbf{r}^k}. \qquad \text{(A.39)}$$

The relations obtained in this Section allow moving to the continuous limit and analyzing the spatial properties of the correlator of non-affine deformations $\mathscr{R}^{\text{na}}$.

Appendix 2

Based on equation (16), $\hat{D}$ is the resolvent for the matrix $\kappa^2 \hat{T}$, and the elements D^{kl} satisfy the following equation:

$$D^{kl} - \kappa^2 \sum_m T^{ml} D^{km} = \delta_{kl}, \qquad \text{(A.40)}$$

where δ_{kl} is the Kronecker symbol.

Let us decompose $D(\mathbf{r}^k, \mathbf{r}^m)$ near the bond l to find the function $D(\mathbf{r}^k, \mathbf{r}^l)$ acting in a continuous representation and depending on the two coordinates of the bonds $\mathbf{r}^k$ and $\mathbf{r}^l$:

$$D(\mathbf{r}^k, \mathbf{r}^m) \simeq D(\mathbf{r}^k, \mathbf{r}^l) + \sum_\alpha D^{(0,1)}_\alpha(\mathbf{r}^k, \mathbf{r}^l)(\mathbf{r}^m - \mathbf{r}^l)_\alpha$$
$$+ \sum_{\alpha,\beta} \frac{D^{(0,2)}_{\alpha\beta}(\mathbf{r}^k, \mathbf{r}^l)}{2} (\mathbf{r}^m - \mathbf{r}^l)_\alpha (\mathbf{r}^{\mathbf{m}} - \mathbf{r}^{\mathbf{l}})_\beta, \qquad \text{(A.41)}$$

where the entry $^{(0,1)}$ and $^{(0,2)}$ denotes taking the first and second derivatives of the corresponding variable. Then

$$\sum_m T^{ml} D(\mathbf{r}^k, \mathbf{r}^m) = D(\mathbf{r}^k, \mathbf{r}^l) \sum_m T^{ml} + \sum_{\alpha,\beta} D^{(0,2)}_{\alpha\beta}(\mathbf{r}^k, \mathbf{r}^l) g^l_{\alpha\beta}, \qquad \text{(A.42)}$$

where the symmetric form of the matrix $\hat{T}$ was taken into account, which follows from its definition (17), resulting in $\sum_m (\mathbf{r}^m - \mathbf{r}^l)_\alpha T^{ml} = 0$. The tensor $g^l_{\alpha\beta}$ is defined as

$$g^l_{\alpha\beta} = \frac{1}{2} \sum_m (\mathbf{r}^m - \mathbf{r}^l)_\alpha (\mathbf{r}^m - \mathbf{r}^l)_\beta T^{ml} \qquad \text{(A.43)}$$

and is a tensor of the second rank. The tensor $g^l_{\alpha\beta}$ is diagonal with equal components in case of an isotropic medium. In addition, when considering a homogeneous medium, when the bonds do not differ from each other, the tensor $g^l_{\alpha\beta}$ does not depend on the number of bond l:

$$g^l_{\alpha\beta} = g \delta_{\alpha\beta}, \quad g = \frac{1}{6K} \sum_{m,l} T^{ml} (\mathbf{r}^m - \mathbf{r}^l)^2. \qquad \text{(A.44)}$$

As follows from the relations (12)−(14),

$$\sum_m T^{ml} = \sum_l T^{ml} = \frac{1-\kappa}{\kappa^2}, \qquad \text{(A.45)}$$

therefore, we have the ratio $g \propto \kappa^{-2}$ for $\kappa \ll 1$.

Thus, taking into account the expressions (A.44) and (A.45) obtained, the equation (A.42) is represented as

$$\sum_m T^{ml} D(\mathbf{r}^k, \mathbf{r}^m) = \frac{(1-\kappa)}{\kappa^2} D(\mathbf{r}^k, \mathbf{r}^l) + g\Delta_{\mathbf{r}^l} D(\mathbf{r}^k, \mathbf{r}^l), \tag{A.46}$$

where $\Delta_{\mathbf{r}^l}$ means the Laplace operator acting on the coordinate $\mathbf{r}^l$ of the bond location l, which in the Cartesian coordinate system $\{\mathbf{e}_x,\ \mathbf{e}_y,\ \mathbf{e}_z\}$ has the form

$$\Delta_{\mathbf{r}^l} = \nabla_{\mathbf{r}^l} \cdot \nabla_{\mathbf{r}^l} = \frac{\partial^2}{\partial (r^l_x)^2}\mathbf{e}_x + \frac{\partial^2}{\partial (r^l_y)^2}\mathbf{e}_y + \frac{\partial^2}{\partial (r^l_z)^2}\mathbf{e}_z. \tag{A.47}$$

Taking into account the found expression (A.46), the equation (A.40) in the continuous limit has the following form:

$$\kappa D(\mathbf{r}^k, \mathbf{r}^l) - g\kappa^2 \Delta_{\mathbf{r}^l} D(\mathbf{r}^k, \mathbf{r}^l) = \delta(\mathbf{r}^k - \mathbf{r}^l), \tag{A.48}$$

where $\delta(\mathbf{r})$ corresponds to the Dirac delta function. This equation for the function $D(\mathbf{r}^k, \mathbf{r}^l)$ has the form of the Helmholtz equation, the solution of which for a three-dimensional system is the following function depending on the coordinate difference $|\mathbf{r}^k - \mathbf{r}^l|$:

$$D(|\mathbf{r}^k - \mathbf{r}^l|) = \frac{e^{-|\mathbf{r}^k - \mathbf{r}^l|/\xi}}{4\pi\kappa\xi^2 |\mathbf{r}^k - \mathbf{r}^l|}, \tag{A.49}$$

where the parameter $\xi = \sqrt{g\kappa}$ defines some spatial scale. Since $g \propto \kappa^{-2}$ for $\kappa \ll 1$, then $\xi \propto \kappa^{-1/2}$.

Similarly, the resolvent $G(\mathbf{r}_i, \mathbf{r}_j)$ is found in a continuous representation as a function of two coordinates of atoms by solving the Poisson equation

$$\Delta G(\mathbf{r}_i, \mathbf{r}_j) = \frac{1}{\kappa\chi}\delta(\mathbf{r}_i - \mathbf{r}_j). \tag{A.50}$$

The following function is the solution to this equation for a three-dimensional system, depending on the difference of coordinates $|\mathbf{r}_i - \mathbf{r}_j|$:

$$G(|\mathbf{r}_i - \mathbf{r}_j|) = -\frac{1}{4\pi\kappa\chi |\mathbf{r}_i - \mathbf{r}_j|}. \tag{A.51}$$

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