Effective local elastic moduli of a strongly disordered medium

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The local elastic properties of amorphous solids are investigated using the theory of correlated random matrices. A significant increase in stiffness is shown in the interfacial region, which characteristic thickness depends on the strength of disorder. The studied interfacial effect increases the influence of nanoparticles on the macroscopic stiffness of nanocomposites. The obtained thickness of the interfacial region has the same order as the length scale of the boson peak.

I. INTRODUCTION

Amorphous glassy materials exhibit spatially inhomogeneous microscopic elastic properties due to their disordered structure [1-4]. The local elastic heterogeneity results in non-affine deformations of amorphous solids under uniform stress. The presence of non-affine deformations was observed in a wide range of amorphous materials: metallic glasses [5], polymer hydrogels [6], supercooled liquids [7], Lennard-Jones glasses [8], and silica glass [9]. The characteristic size of non-affine deformations was estimated as tens of particle sizes for Lennard-Jones glasses [10]. For smaller length scales, the classical continuum elasticity theory can not be applied [11].

If the size of an amorphous medium is much larger than its non-affine length scale, one can use the macroscopic elastic moduli to describe the mechanical properties of this system. However, in composite systems containing amorphous materials, some regions may have small typical sizes. An important example is nanocomposites, in which the size of nano-inclusions may be comparable to the non-affine length scale of the host amorphous medium. Therefore, it is important to study the local elastic properties of amorphous solids, especially near the interface with other materials.

Amorphous polymers are an important class of amorphous materials. The elastic properties of polymer nanocomposites attract considerable interest due to their unique properties and great potential as future materials [12-15]. It was established that polymer doping with nanoparticles even in small concentrations could lead to significant changes in the elasticity of the host material [16-20].

It was proposed that the elastic properties of nanocomposites can be described by the so-called three-phase model [21]. The model assumes that the structure of a polymer is perturbed around the nanoparticle, which results in an effective interphase region around the nanoparticle with intermediate elastic properties. The interphase region has a strong influence on the macroscopic stiffness of the nanocomposite due to the large total surface area of nanoparticles. At present, the three-phase model is usually used as a phenomenologic model to fit the influence of inclusions on macroscopic elastic moduli obtained experimentally or using molecular dynamics [22-27].

Recent molecular dynamics studies have directly shown an increase in local elastic moduli of epoxy near the boehmite nanolayer [28] and polystyrene near the silica nano-inclusion [29]. In the latter case, an increase in polystyrene stiffness was revealed within a characteristic range of 1.4 nm from the nanoparticle, while polystyrene density saturates to the bulk value at significantly shorter distances. The enhancement of the local elastic properties of the polymer was attributed to the effect of non-affine deformations, which requires a more detailed theoretical study.

It was shown that general vibrational and mechanical properties of amorphous solids can be studied by the random matrix model [30, 31]. Recently, using the theory of correlated random matrices, the analytical form of the vibrational density of states and the dynamical structure factor was obtained [32].

In the present paper the theory of the correlated random matrices is applied to study the effect of disorder on local elastic properties.

II. LINEAR RESPONSE

Macroscopic elastic properties determine the relationship between the average strain of a system and the stress applied to the system. In this paper, the linear elastic properties are considered. In the general case, a linear response to some external force at frequency \( \omega \) is determined by the following equation

\[
\sum_j [\Phi_{ij} - \omega^2 m_{ij}] u_j = f_i, \tag{1}
\]

where \( \Phi \) is the force constant matrix, \( \hat{m} \) is the mass matrix, \( u_j \) is the displacement of \( j \)th degree of freedom from the equilibrium position, and \( f_i \) is a force applied to \( i \)th degree of freedom.

In an amorphous system, the force-constant matrix \( \Phi \) has a random nature. At the same time, the mass matrix \( \hat{m} \) is non-random. Usually, it is diagonal, but in this paper a general non-random matrix \( \hat{m} \) is considered.

For each realization of the force-constant matrix \( \Phi \), the response \( u_i \) is different. However, the fluctuations of macroscopic quantities based on \( u_i \) (e.g. the macroscopic strain) have negligible fluctuations. Therefore, the average reaction \( \langle u_i \rangle \) can be considered, where the angle brackets denote the averaging over different realizations of \( \Phi \). To analyze the average response, the
For a strongly disordered system, the matrix $\hat{\Phi}$ is introduced, where the average response can be written as

$$\langle u_i \rangle = -\sum_j G_{ij}(\omega^2) f_j.$$  

(3)

The resolvent $\hat{G}(z)$ can be presented as

$$\hat{G}(z) = \frac{1}{nz - \hat{\Phi} \text{eff}(z)},$$  

(4)

where $\hat{\Phi} \text{eff}(z)$ is an effective force constant matrix, which gives the same average response as the random force constant matrix $\hat{\Phi}$:

$$\sum_j [\hat{\Phi} \text{eff}(\omega^2) - \omega^2 m_{ij}] \langle u_j \rangle = f_i.$$  

(5)

For a strongly disordered system, the matrix $\hat{\Phi} \text{eff}(z)$ may differ significantly from the mean force constant matrix $\langle \hat{\Phi} \rangle$.

### III. RANDOM MATRIX MODEL

Near the stable equilibrium, the force-constant matrix $\hat{\Phi}$ is positive definite. The condition of positive definiteness is equivalent to the possibility to represent the matrix $\hat{\Phi}$ in the form

$$\hat{\Phi} = \hat{A} \hat{A}^T,$$  

(6)

where $\hat{A}$ is some rectangular matrix. The $i$th row of the matrix $\hat{A}$ corresponds to the $i$th degree of freedom. The $k$th column corresponds to the $k$th bond, which has the non-negative quadratic potential energy

$$U_k = \frac{1}{2} \left( \sum_i A_{ik} u_i \right)^2.$$  

(7)

Each bond may involve several degrees of freedom depending on the number of non-zero elements in $k$th column of the matrix $\hat{A}$. In this paper, a system with $N$ degrees of freedom and $K$ bonds will be considered, which corresponds to $N \times K$ matrix $\hat{A}$.

For amorphous solids, the matrix $\hat{A}$ has a random nature. The $k$th column of the matrix $\hat{A}$, which corresponds to the $k$th bond, may have its own correlation matrix $C^{(k)}_{ij} = \langle A_{ik} A_{jk} \rangle$. At the same time, different columns of the matrix $\hat{A}$ representing different bonds are assumed to be uncorrelated with each other. So the pair correlations of the elements of the matrix $\hat{A}$ have the form

$$\langle A_{ik} A_{jl} \rangle = C^{(k)}_{ij} \delta_{kl}.$$  

(8)

The number of nonzero elements in the matrix $\hat{C}^{(k)}$ depends on the number of degrees of freedom, which are involved in $k$th bond. In the previous paper, the difference between correlation matrices $\hat{C}^{(k)}$ was not taken into account. However, different bonds have different positions in space and involve different sets of degrees of freedom, which plays a crucial role for local elastic properties.

In the assumption that elements of the matrix $\hat{A}$ are Gaussian random numbers with zero mean, the random matrix theory can be applied. The method described in [32] can be generalized to the case in which each column of the matrix $\hat{A}$ has its own correlation matrix $C^{(k)}$. The resulting effective force constant matrix is

$$\hat{\Phi} \text{eff}(z) = \sum_k E_k(z) \hat{C}^{(k)},$$  

(9)

where

$$E_k(z) = \frac{1}{1 - \text{Tr} [\hat{C}^{(k)} \hat{G}(z)]}.$$  

(10)

Using the definition of $\hat{G}(z)$, the system of nonlinear equations for coefficients $E_k(z)$ is obtained:

$$E_k(z) = 1 + \text{Tr} \left[ E_k(z) \hat{C}^{(k)} \frac{1}{nz - \sum_l E_l(z) \hat{C}^{(l)}} \right].$$  

(11)

In a general case, Eq. (11) can be solved numerically for any set of correlation matrices $\hat{C}^{(k)}$. However, in some cases Eq. (11) can be simplified, which is considered below.

### IV. EFFECTIVE ELASTIC MEDIUM

In this section an amorphous solid with homogeneous statistical properties is considered. For a such medium, one can assume a homogeneous distribution of $K$ bonds over a system with $N$ degrees of freedom. In this case one can introduce a smooth function $E(r, z)$ such that $E_k(z) = E(r_k, z)$, where $r_k$ is a coordinate of $k$th bond.

In the bulk of amorphous solid, $E(r, z)$ does not depend on the coordinate $r$. However, the boundary effects may lead to non-homogeneous $E(r, z)$ near the boundaries of amorphous solids. In Appendix A the differential equation for $E(r, z)$ is derived. In the static case ($z = \omega^2 = 0$), the equation for $E(r) \equiv E(r, 0)$ reads as

$$(1 + \varkappa) E(r) = \varkappa + \xi_0^2 \Delta \ln E(r),$$  

(12)

where $\varkappa = K/N - 1$, $\xi_0$ is a typical bond size, and $\Delta$ denotes the Laplacian.

For slowly varying $E(r)$ in the region near the point $r$, the effective dynamical matrix has the form $\hat{\Phi} \text{eff} = E(r) \langle \hat{\Phi} \rangle$, where $\langle \hat{\Phi} \rangle = \sum_k \hat{C}^{(k)}$ is the averaged force constant matrix. Therefore, $E(r)$ can be considered
as a dimensionless elasticity since elastic moduli of the reference medium described by \( \Phi \) are multiplied by \( E(r) \).

In the bulk of amorphous solid \( E(r) = E_\infty = \kappa/(\kappa + 1) \). If the number of random bonds is much greater than the number of degrees of freedom \( K \gg N \), self-averaging of random bonds takes place. It results in small fluctuations of the force constant matrix \( \Phi \). In this case \( \Phi^{\text{eff}} \) is close to the average force constant matrix \( \langle \Phi \rangle \). The role of disorder controlled by the parameter \( \kappa \) on the vibrational properties of the bulk amorphous solid was studied in [32].

Equation (12) can be written as

\[
\frac{E(r)}{E_\infty} = 1 + \xi^2 \Delta \ln \frac{E(r)}{E_\infty},
\]

where \( \xi = \xi_b/\sqrt{\kappa} \) is the only dimensional parameter in the above equation. Therefore, \( \xi \) defines the length scale of the boundary effects. Since \( \xi \approx \kappa^{-1/2} \), it depends on the strength of disorder. For strongly disordered medium \( \xi \gg \xi_b \).

To obtain \( E(r) \) in the whole amorphous solid, Eq. (13) should be accomplished with the boundary conditions. The most important case is the interface of amorphous medium with a more rigid and ordered medium. Such a rigid and ordered medium can be simulated as a medium with \( \kappa \gtrsim 1 \). In this case, one can assume \( E(r) \approx 1 \) on the boundaries.

For strongly disordered medium \( \kappa \ll 1 \), the boundary condition is \( E(r)/E_\infty \approx 1/\kappa \gg 1 \). Without the loss of precision, one can assume that \( E(r)/E_\infty = \infty \) on the boundaries to solve Eq. (13). Below, the two most important geometries of the boundary of an amorphous body are considered.

### A. Flat boundary

Near a flat boundary, \( E(r) \) depends only on the distance from the boundary, which is denoted by \( x \). In this case, Eq. (13) has the one-dimensional form

\[
\frac{E(x)}{E_\infty} = 1 + \xi^2 \Delta \ln \frac{E(x)}{E_\infty}. \tag{14}
\]

The solution of Eq. (14) has a universal dependence on the scaled coordinate \( x/\xi \), which is shown in Fig. 1. Far away from the boundary \( (x \gg \xi) \), the asymptotic solution is

\[
E(x) = E_\infty \left( 1 + c_1 e^{-x/\xi} \right), \tag{15}
\]

where \( c_1 \approx 2.5527 \). Near the boundary \( (x \ll \xi) \), the asymptotic solution is

\[
E(x) = E_\infty \frac{2\xi^2}{x^2}. \tag{16}
\]

### B. Spherical inclusion

Another important example is spherical nanoinclusions in an amorphous medium. Around each nanoinclusions, Eq. (13) can be written in spherical coordinates

\[
\frac{E(r)}{E_\infty} = 1 + \xi^2 \frac{\partial}{r^2 \partial r} \left( r^2 \frac{\partial}{\partial r} \ln \frac{E(r)}{E_\infty} \right), \tag{17}
\]

where \( r \) is the distance from the center of the nanoinclusion of the radius \( R \). The solution of Eq. (17) is shown in Fig. 2. Far away from the nanoinclusion \( (r \gg \xi) \), the asymptotic solution is

\[
E(r) = E_\infty \left( 1 + c_2 \frac{R}{r} e^{-(r-R)/\xi} \right), \tag{18}
\]
where $c_2$ is a coefficient, which depends on the ratio $R/\xi$. The asymptotic solution near the surface of nanoinclusion $(r - R \ll \xi, R)$ is

$$E(r) = E_\infty \frac{2R^2\xi^2}{r^2(r - R)^2}. \quad (19)$$

As in the one-dimensional case, the solution $E$ is inapplicable in a thin near-boundary region $r - R \lesssim \xi$. Thus, the actual near-boundary value of $E(r)$ is $E(R + \xi_b) \sim 1$.

V. DISCUSSION

The obtained effective force constant matrix $\Phi_{\text{eff}}$ can be used as a non-random substitution of the random force constant matrix $\Phi$, which gives the same average response to external forces. Particularly, $\Phi_{\text{eff}}$ can be used to represent the macroscopic elastic properties of composite materials containing amorphous materials.

The same effective force constant matrix $\Phi_{\text{eff}}$ can be used to find the average response to microscopical forces at any length scale. At the same time, one can expect strong fluctuations of atomic displacements for length scales below $\xi$. The analysis of fluctuations is the subject of the future work.

Using the random matrix theory, the effective force constant matrix $\Phi_{\text{eff}}$ was obtained as a sum of correlation matrices $C^{(k)}$ with some coefficients defined by Eq. (11). In amorphous system, bonds usually have a finite range, which results in sparse matrices $C^{(k)}$. The results of the random matrix are applicable if the number of nonzero elements is much bigger than one. This assumption works reasonably well, since the interaction between atoms involves two or more atoms, each of which has three degrees of freedom. Additionally, the numerical random matrix model described in [32] was analyzed and compared with the present theory. The effective force constant matrix obtained numerically quickly converges to the present theoretical predictions with the increase of the radius of bonds.

Equation (19) shows that the effective force constant matrix $\Phi_{\text{eff}}$ describes a short-range interaction if matrices $C^{(k)}$ describes finite-range bonds. This property holds for any strength of disorder, so the effective medium described by $\Phi_{\text{eff}}$ can be analyzed using the continuum elasticity theory.

For amorphous solids with homogeneous and isotropic statistics, Eq. (13) defines the static local stiffness of the effective medium. Near the boundary with a more rigid medium, the static local stiffness of the effective medium exceeds its bulk values. The length scale $\xi \sim \kappa^{-1/2}$ defines the thickness of the boundary layer with increased stiffness.

The studied effect is especially important for nanocomposites. For homogeneous host material, the macroscopic stiffness of the nanocomposite can be calculated using the Mori-Tanaka approach [36, 37]. The addition of a small concentration of rigid spherical inclusions to the host material leads to the following relative increase of the bulk and shear modulus, respectively:

$$\frac{K_{\text{nc}} - K_0}{K_0} = 4\pi nR^3 \frac{1 - \nu}{1 + \nu}, \quad (20)$$

$$\frac{\mu_{\text{nc}} - \mu_0}{\mu_0} = 10\pi nR^3 \frac{1 - \nu}{4 - 5\nu}, \quad (21)$$

where $n$ is the concentration of nanoinclusions, $R$ is the radius of nanoinclusions, $\nu$ is the Poisson ratio of the host material. The disorder of the host material leads to the formation of the effective shell of the thickness $\xi$ with increased stiffness around each nanoparticle. In this case, the nanoparticles have the effective radius $R_{\text{eff}}$ such that $R_{\text{eff}} - R \sim \xi$. Thus, for $R \sim \xi$ the influence of nanoinclusions on the macroscopic stiffness of the nanocomposite will be increased by an order of magnitude.

It is important that the studied effect is determined by the strength of the disorder. This effect should be distinguished from the effect of adhesion, which can directly change the density and other structural properties of amorphous medium near the surface of nanoinclusions.

The main result [13] concerns the static stiffness, however, Eqs. (11) and (A6) may be applied to arbitrary frequency $\omega$ given by the parameter $z = \omega^2$. For amorphous solids with homogeneous and isotropic statistics, far away from boundaries $E_k(z) = E_\infty(z)$. In this case Eq. (11) can be written as a complex equation

$$\kappa Z + \frac{Z^2}{N} \text{Tr} \left[ \frac{1}{Z - \langle M \rangle} \right] = (1 + \kappa)z, \quad (22)$$

where $Z = z/E_\infty(z)$ and $\langle M \rangle = \langle \Phi \rangle \bar{m}^{-1/2}$ is the average dynamical matrix. Thus, for any given $z = \omega^2 - i0$ one can find $Z(z)$ and $E_\infty(z) = z/Z(z)$. Using the solution $Z(z)$, the vibrational density of states $g(\omega) = (1 + \kappa)\frac{2\pi}{\omega} \text{Im}[1/Z(z^2 - i0)]$ and other vibrational properties can be analyzed. A more detailed analysis of vibrational properties has been done in [32].

Amorphous solids have an excess of low-frequency vibrational density of states, known as the boson peak [38, 39]. The boson peak length scale defined as $\xi_{\text{bp}} = 2\pi c_T/\omega_{\text{bp}}$, where $c_T$ is the transverse sound velocity and $\omega_{\text{bp}}$ is the boson peak frequency, was attributed to the heterogeneity length scale [10]. In the random matrix model, the boson peak length scale is $\xi_{\text{bp}} \sim a_0 \kappa^{-1/2}$, where $a_0$ is the interatomic distance [32]. Thus, the length scales $\xi$ and $\xi_{\text{bp}}$ have the same order and the same dependence on the strength of disorder in the studied random matrix model.

In real amorphous solids, the strength of disorder cannot be varied in a wide range. However, model granular systems, known as jammed solids, have the possibility to change their properties significantly [10]. This is due to a critical behavior of elastic and vibrational properties for small positive values of the
parameter \( z - z_0 \) \([11,13]\), which corresponds to the parameter \( \zeta \) in the present theory \([32]\). In jammed solids, the length scale \( l_0 \sim (z - z_0)^{-1/2} \) is related to the breakdown of the continuum elasticity \([11]\) and coincides with the boson peak length scale \([11]\). Thus, the length scale \( l_0 \) corresponds to the length scale \( \xi \) in the present theory. The investigation of the local elastic properties near the boundaries of jammed solids is of great interest to check the validity of Eq. \([13]\) for such systems.

The obtained results are not limited to the study of the elastic properties of strongly disordered systems. Other properties requiring positive definiteness can be considered. For example, instead of stiffness, one can consider the conductivity of a strongly disordered medium. Thus, \( E(\mathbf{r}) \) can describe the increase in the conductivity of the effective medium near the interface with a well-conducting material. However, the applicability of the considered model to such kind of systems requires further research.

VI. CONCLUSION

In this paper, the theory of the correlated random matrices was applied to find the local elastic properties of amorphous solids. The effective force constant matrix \( \Phi \) was obtained, which can be used to find the average linear response to a force of a given frequency \( \omega \) given by the parameter \( z = \omega^2 \).

For amorphous solid with homogeneous and isotropic statistical properties, a continuous equation for static stiffness \( E(\mathbf{r}) \) was obtained. It reveals the increase of the stiffness of amorphous solid near the boundary with a more rigid and ordered body. The typical thickness of the boundary layer with increased stiffness is \( \xi \sim \zeta^{-1/2} \). Far away from the boundaries \( E(\mathbf{r}) \) has an exponential decay to the bulk value \( E_\infty \) with a typical length \( \xi \).

For the strongly disordered amorphous solids \( \zeta \ll 1 \), the length scale \( \xi \) is much larger than the typical interatomic size in the system. The scaling of \( \xi \) with the strength of disorder emphasizes the role of disorder in the formation of the boundary layer with increased stiffness.

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Appendix A: Amorphous solid with homogeneous and isotropic statistical properties

One can assume that \( E_i(z) = E(\mathbf{r}_1, z) \) is close to \( E_k(z) = E(\mathbf{r}_k, z) \) for neighbor bonds \( k \) and \( l \). In this case, Eq. \((11)\) can be written as

\[
E_k(z) = 1 + W_k(Z) + \sum_l W_{kl}(Z) \frac{E_l(z) - E_k(z)}{E_k(z)} + \sum_{lm} W_{klm}(Z) \frac{(E_l(z) - E_k(z))(E_m(z) - E_k(z))}{E_k^2(z)},
\]

where \( Z = z/E(z) \) and

\[
W_k(Z) = \text{Tr} \left[ \hat{C}^{(k)} \frac{1}{\hat{m}Z - \langle \Phi \rangle} \right], \tag{A2}
\]

\[
W_{kl}(Z) = \text{Tr} \left[ \hat{C}^{(k)} \frac{1}{\hat{m}Z - \langle \Phi \rangle} \hat{C}^{(l)} \frac{1}{\hat{m}Z - \langle \Phi \rangle} \right], \tag{A3}
\]

\[
W_{klm}(Z) = \text{Tr} \left[ \hat{C}^{(k)} \frac{1}{\hat{m}Z - \langle \Phi \rangle} \hat{C}^{(l)} \frac{1}{\hat{m}Z - \langle \Phi \rangle} \hat{C}^{(m)} \frac{1}{\hat{m}Z - \langle \Phi \rangle} \right]. \tag{A4}
\]

At the same time, \( E_i(z) - E_k(z) \) can be written as

\[
E_i(z) - E_k(z) = \sum_{\alpha} \frac{\partial E(\mathbf{r}_k, z)}{\partial r_{\alpha}} (r_{\alpha} - r_{\beta}) + \frac{1}{2} \sum_{\alpha\beta} \frac{\partial^2 E(\mathbf{r}_k, z)}{\partial r_{\alpha} \partial r_{\beta}} (r_{\alpha} - r_{\beta})(r_{\beta} - r_{\gamma}). \tag{A5}
\]

As a result, the following differential equation for \( E(\mathbf{r}, z) \) is obtained:

\[
E(\mathbf{r}, z) = 1 + W(\mathbf{r}, Z) + \frac{1}{E(\mathbf{r}, z)} \sum_{\alpha} W'_\alpha(\mathbf{r}, Z) \frac{\partial E(\mathbf{r}, z)}{\partial r_{\alpha}} + \frac{1}{E(\mathbf{r}, z)} \sum_{\alpha\beta} W'_{\alpha\beta}(\mathbf{r}, Z) \frac{\partial^2 E(\mathbf{r}, z)}{\partial r_{\alpha} \partial r_{\beta}} + \frac{1}{E(\mathbf{r}, z)^2} \sum_{\alpha\beta} W''_{\alpha\beta}(\mathbf{r}, Z) \frac{\partial E(\mathbf{r}, z)}{\partial r_{\alpha}} \frac{\partial E(\mathbf{r}, z)}{\partial r_{\beta}}, \tag{A6}
\]

where

\[
W(\mathbf{r}_k, Z) = W_k(Z), \tag{A7}
\]

\[
W'_\alpha(\mathbf{r}_k, Z) = \sum_{l} W_{kl}(Z) (r_{\alpha} - r_{\beta}), \tag{A8}
\]

\[
W'_{\alpha\beta}(\mathbf{r}_k, Z) = \frac{1}{2} \sum_{l} W_{kl}(Z) (r_{\alpha} - r_{\beta})(r_{\beta} - r_{\gamma}), \tag{A9}
\]

\[
W''_{\alpha\beta}(\mathbf{r}_k, Z) = \sum_{lm} W_{klm}(Z) (r_{\alpha} - r_{\beta})(r_{\beta} - r_{\gamma}). \tag{A10}
\]
Static properties are defined by the limit $z \to 0$ and $Z \to 0$. In this case there are the following sum rules:

$$\sum_k W_k(0) = N_0 - N, \quad (A11)$$
$$\sum_i W_{ki}(0) = -W_k(0), \quad (A12)$$
$$\sum_m W_{klm}(0) = -W_{kl}(0), \quad (A13)$$

where $N_0$ is a number of trivial zero-frequency modes (translations and rotations), which can be neglected for $N \gg 1$. Using Eqs. (A11)–(A13), for an amorphous solid with homogeneous and isotropic statistical properties, one obtain

$$W(r_k, 0) = -\frac{N}{K}, \quad (A14)$$
$$W''(r_k, 0) = 0, \quad (A15)$$
$$W_{\alpha\beta}(r_k, 0) = \frac{N}{K} \xi^2 \delta_{\alpha\beta}, \quad (A16)$$
$$W''_{\alpha\beta}(r_k, 0) = -\frac{N}{K} \xi^2 \delta_{\alpha\beta}, \quad (A17)$$

where $\xi_k$ is a typical radius of the bonds.

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