

Analysis of spatial correlations in a model two-dimensional liquid through eigenvalues and eigenvectors of atomic-level stress matrices

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Considerations of local atomic-level stresses associated with each atom represent a particular approach to address structures of disordered materials at the atomic level. We studied structural correlations in a two-dimensional model liquid using molecular dynamics simulations in the following way. We diagonalized the atomic-level stress tensor of every atom and investigated correlations between the eigenvalues and orientations of the eigenvectors of different atoms as a function of distance between them. It is demonstrated that the suggested approach can be used to characterize structural correlations in disordered materials. In particular, we found that changes in the stress correlation functions on decrease of temperature are the most pronounced for the pairs of atoms with separation distance that corresponds to the first minimum in the pair density function. We also show that the angular dependencies of the stress correlation functions previously reported by Wu *et al.* [*Phys. Rev. E* **91**, 032301 (2015)] do not represent the anisotropic Eshelby's stress fields, as it is suggested, but originate in the rotational properties of the stress tensors.

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I. INTRODUCTION

It is relatively easy to describe structures of crystalline materials due to the presence of translational periodicity. This periodicity implies that atoms whose coordinates differ by a vector of translation would have identical atomic environments, if all atoms were at equilibrium positions. In glasses and liquids, in contrast, there is no translational periodicity and every atom, in principle, can have a distinct atomic environment [1,2]. Largely for this reason the description of disordered materials continues to be a challenge. Many different approaches have been suggested to describe disordered structures. However, none of them allows establishing a clear link between the structural and dynamic properties of disordered matter [1,2].

The concept of local atomic-level stresses was introduced to describe model structures of metallic glasses and their liquids [2–6]. For a particle i surrounded by particles j , with which it interacts through pair potential $U(r_{ij})$, the $\alpha\beta$ component of the atomic-level stress tensor of atom i is defined as [3–6]:

$$\sigma_i^{\alpha\beta} = \frac{1}{V_i} \sum_{j \neq i} \left[\frac{dU}{dr_{ij}} \right] \left(\frac{r_{ij}^\alpha r_{ij}^\beta}{r_{ij}} \right). \quad (1)$$

The sum over j in (1) is over all particles with which particle i interacts. In (1) V_i is the local atomic volume. In the present considerations it is assumed that V_i is just the inverse of the average atomic number density $V_i \equiv 1/\rho_0$. By convention, the definition without V_i corresponds to the local atomic-level stress element [7,8]. Note that the α component of the force acting on particle i from particle j is $f_{ij}^\alpha = [dU(r_{ij})/dr_{ij}](r_{ij}^\alpha/r_{ij})$, where $\vec{r}_{ij} = \vec{r}_j - \vec{r}_i$ is the radius vector from i to j . Also note that the atomic-level stress tensor (1) is symmetric with respect to the indexes α and β . Thus in three dimensions it has six independent components [4–6], while in two dimensions it has three independent components.

There are several important results associated with the concept of atomic-level stresses. One result is the equipartition of the atomic-level stress energies in liquids [4–6]. Thus the energies of the atomic-level stress components were defined and it was demonstrated for the studied model liquid systems in three dimensions that the energy of every stress component is equal to $k_b T/4$. Thus the total stress energy, which is the sum of the energies of all six components, is equal to $6 \times k_b T/4 = (3/2)k_b T$, i.e., the potential energy of a classical three-dimensional (3D) harmonic oscillator. An explanation for this result has been suggested [4–6]. The equipartition breaks down in the glass state. Then there was an attempt to describe the glass transition and fragilities of liquids on the basis of atomic-level stresses [9]. Another result is related to the Green-Kubo expression for viscosity. Thus the correlation function between the macroscopic stresses that enter into the Green-Kubo expression for viscosity was decomposed into the correlation functions between the atomic-level stress elements. Considerations of the obtained atomic-level correlation functions allowed demonstration of the relation between the propagation and dissipation of shear waves and viscosity. This result, after all, is not surprising in view of the existing generalized hydrodynamics and mode-coupling theories [10,11]. However, in Refs. [7,8,12,13] the issue has been addressed from a new perspective and the relation between viscosity and shear waves was demonstrated very explicitly.

Recently it has been claimed in Ref. [14] that considerations of the correlations between the atomic-level stresses allow observation of the angular-dependent stress fields, which are present in liquids in the absence of any external shear. In many respects our attempt to understand the results presented in Ref. [14] lead to the present publication.

Here we demonstrate that the angular dependencies of the stress correlation functions presented in Ref. [14] do not correspond to the angular-dependent stress fields (which can exist in the system). We show that the angular dependencies of

the stress correlation functions observed in Ref. [14] originate from the rotational properties of the stress tensors.

However, the ideas presented here go beyond the scope of Ref. [14]. Here we address the atomic-level stresses and correlations between the atomic-level stresses of atoms separated by some distance from a new and yet very natural perspective. It is surprising that this approach has not been investigated in detail before. Reasoning in a similar direction was presented in Refs. [15,16]. However, considerations presented there do not address correlations between the atomic-level stresses of different atoms.

This paper is organized as follows. In Sec. II the idea of the approach is presented. Section III is a reminder about transformational properties of the stress tensors. Atomic-level stress correlations functions are discussed in the context of the present approach in Sec. IV. In Sec. V the connection between the Eshelby's inclusion problem and the atomic-level stress correlation functions is analyzed. In Sec. VI the results of our MD simulations are described. We conclude in Sec. VII.

II. STRESS TENSOR ELLIPSES

The atomic-level stress tensor $\sigma_i^{\alpha\beta}$ defined with Eq. (1) is real and symmetric. Thus it can be diagonalized and, in two dimensions, two real eigenvalues (λ_i^1 and λ_i^2) and two real eigenvectors can be found. The tensor $\sigma_i^{\alpha\beta}$ in two dimensions has three independent components. These three parameters determine two eigenvalues and the rotation angle that describes orientation of the orthogonal eigenvectors with respect to the reference coordinate system. Let us associate with each atom i an ellipse with principal axes oriented along the eigenvectors of $\sigma_i^{\alpha\beta}$ and having lengths λ_i^1 and λ_i^2 , as depicted in Fig. 1.

Previously atomic-level stresses were discussed mostly in three dimensions. In 3D symmetric atomic-level stress tensors have six independent components. Thus, previously, in particular in discussions related to the atomic-level stress energies, it was assumed that the local atomic environment of an atom is described by six independent stress components. However, in view of the present considerations, it is clear that

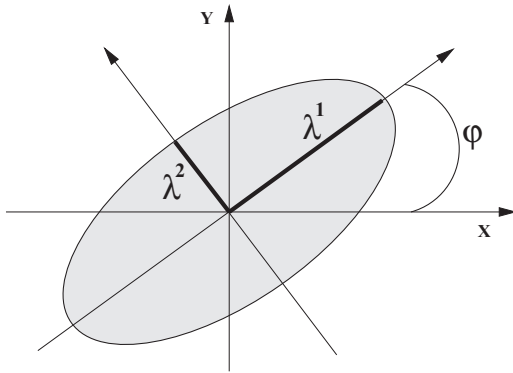


FIG. 1. Atomic-level stress tensor of any atom can be diagonalized. Obtained eigenvalues, λ^1 and λ^2 , can be associated with the lengths of the principal ellipse's axes. The orientation of the ellipse with respect to the reference coordinate system is given by the angle, φ , between the longest ellipse's axis and the x axis of the reference frame.

if the atomic-level stress tensor is diagonalized then its three eigenvalues describe the geometry of local atomic environment, while its three eigenvectors describe the orientation of the associated ellipsoid with respect to the chosen coordinate system.

In model metallic glasses in three dimensions atoms often have 12 or 13 nearest neighbors [1,2,17]. Working with atomic-level stresses effectively reduces the richness of all possible local atomic geometries to just three numbers. Of course, that is more convenient than dealing with more numbers associated, for example, with the description based on Voronoi indexes [1,2]. However, it is unclear for which purposes it is enough to consider only three numbers and for which purposes it may not be enough. In consideration of the stress correlations between two atoms in three dimensions there are 12 physically relevant parameters: six eigenvalues (three on each atom) describe the geometries of the two ellipsoids and six parameters describe orientations of the ellipsoids with respect to the line from one ellipsoid to another. A representation in a particular coordinate frame needs another three parameters that describe the orientation of the line from one atom to another.

In Refs. [15,16] correlations between the eigenvalues of the same atom has been considered for 2D and 3D Lennard-Jones liquids. There it was argued that there are correlations between the stress eigenvalues of the same atom.

Here we are interested in the correlations between the stress elements of different atoms. If we want to consider stress correlations between two different atoms in two dimensions then we associate ellipses with both atoms and consider the correlations between the eigenvalues and the orientations of the ellipses, see Fig. 2. It is clear that in isotropic one-component liquids all physically meaningful pair correlation functions should depend only on distance r_{ij} .

If the local atomic-level stress tensor is known in the reference frame then its eigenvalues and eigenvectors can be found. In two dimensions we have:

$$\lambda_i^{1,2} = (1/2)[(\sigma_i^{xx} + \sigma_i^{yy}) \pm \sqrt{(\sigma_i^{xx} - \sigma_i^{yy})^2 + 4(\sigma_i^{xy})^2}] \quad (2)$$

$$\tan(\varphi_i^{1,2}) = (V_{i,y}^{1,2}/V_{i,x}^{1,2}) = \sigma_i^{xy}/(\lambda_i^{1,2} - \sigma_i^{yy}). \quad (3)$$

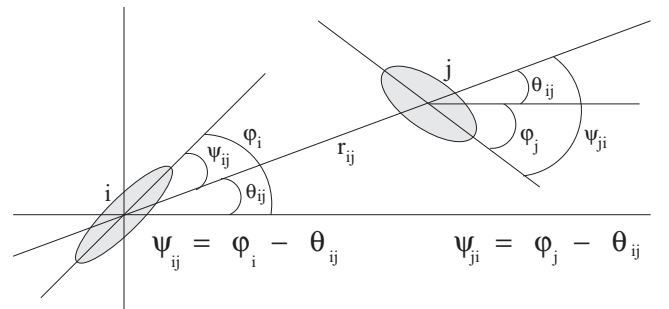


FIG. 2. The orientations of the ellipses with respect to the line connecting atoms i and j are given by the angles ψ_{ij} and ψ_{ji} . The orientation of the line connecting atoms i and j with respect to the X axis of the reference frame is given by the angle θ_{ij} .

Further we will assume that $\varphi_i \in (-\pi/2, \pi/2]$. Physically angle φ_i is defined up to an integer multiple of π as rotation by angle π does not change the ellipse.

In the orthogonal coordinate system based on the eigenvectors of a particular local atomic stress tensor this stress tensor is diagonal with eigenvalues λ^1 and λ^2 on the diagonal. In potentials with repulsive and attractive parts the values of some λ can be negative. The negative value of λ corresponds to the case when the atomic environment of an atom is dilated along the eigenvector associated with this λ . Here we assume that potentials that we consider are purely repulsive. Such systems are held together at some density by periodic boundary conditions. In such cases, both λ^1 and λ^2 are positive and we order them to have $\lambda^1 \geq \lambda^2$.

III. TRANSFORMATIONS OF STRESS TENSORS UNDER ROTATIONS

In this section we provide some well-known facts about transformations of stress tensors under rotations [18]. We will need these facts in our further considerations.

Let us suppose that there are A and B coordinate frames in two dimensions and that frame B is rotated with respect to frame A on angle θ in the counterclockwise direction. The components of the stress tensor S in frame B can be expressed through the components of the stress tensor in frame A using the rotation matrix $R(\theta)$:

$$S_B = R(\theta)S_A R^T(\theta), \quad R(\theta) \equiv \begin{bmatrix} \cos(\theta) & \sin(\theta) \\ -\sin(\theta) & \cos(\theta) \end{bmatrix}, \quad (4)$$

where $R^T(\theta)$ is the transpose of $R(\theta)$. In terms of components, (4) leads to:

$$\sigma_B^{xx} = \sigma_A^{xx} [\cos(\theta)]^2 + \sigma_A^{yy} [\sin(\theta)]^2 + \sigma_A^{xy} \sin(2\theta), \quad (5)$$

$$\sigma_B^{yy} = \sigma_A^{xx} [\sin(\theta)]^2 + \sigma_A^{yy} [\cos(\theta)]^2 - \sigma_A^{xy} \sin(2\theta), \quad (6)$$

$$\sigma_B^{xy} = -(1/2)[\sigma_A^{xx} - \sigma_A^{yy}] \sin(2\theta) + \sigma_A^{xy} \cos(2\theta). \quad (7)$$

Let us now suppose that the angle between the first eigenvector of atom i and the \hat{x} axis of our reference frame is φ_i . In the frame of its eigenvectors the components of the stress tensor of atom i are $\sigma_i^{xx} = \lambda_i^1$, $\sigma_i^{yy} = \lambda_i^2$, $\sigma_i^{xy} = 0$, $\sigma_i^{yx} = 0$. In order to find the components of the stress tensor of atom i in our reference frame we should rotate the components of the stress tensor in the frame of its eigenvectors on angle $-\varphi_i$ using (5), (6), (7). Thus we get:

$$\sigma_i^{xx} = \lambda_i^1 \cos^2(\varphi_i) + \lambda_i^2 \sin^2(\varphi_i), \quad (8)$$

$$\sigma_i^{yy} = \lambda_i^1 \sin^2(\varphi_i) + \lambda_i^2 \cos^2(\varphi_i), \quad (9)$$

$$\sigma_i^{xy} = (1/2)(\lambda_i^1 - \lambda_i^2) \sin(2\varphi_i). \quad (10)$$

Note also that: $\sigma_i^{xx} - \sigma_i^{yy} = (\lambda_i^1 - \lambda_i^2) \cos(2\varphi_i)$.

IV. CORRELATION FUNCTIONS BETWEEN THE ELEMENTS OF ATOMIC-LEVEL STRESS TENSORS OF DIFFERENT ATOMS

In this section we derive the expressions for selected correlation functions between the atomic-level stress elements in terms of eigenvalues and eigenvectors of atomic-level stress matrices.

A. Correlation functions in the directional frame

It is useful to start this section from an argument that plays a very important role in this paper.

Let us consider a pair of atoms i and j separated by radius vector $\mathbf{r}_{ij} = \mathbf{r}_j - \mathbf{r}_i$. We associate with the direction of \mathbf{r}_{ij} a directional coordinate \mathbf{r}_{ij} frame whose \hat{x} axis is along \mathbf{r}_{ij} . The notations $\sigma_{ij}^{\alpha\beta}(i)$ and $\sigma_{ij}^{\delta\gamma}(j)$ will be used for the $\alpha\beta$ and $\delta\gamma$ components of the stress tensors of atoms i and j in the \mathbf{r}_{ij} frame. Further, we consider the products $\sigma_{ij}^{\alpha\beta}(i)\sigma_{ij}^{\delta\gamma}(j)$ in the \mathbf{r}_{ij} -directional frame and average such products over the pairs of atoms separated by radius vector $\mathbf{r}_{ij} = \mathbf{r}$. It is important that this averaging is performed over the values of the stress tensor components in the representation associated with the \mathbf{r}_{ij} frame.

For the following it is necessary to realize that for isotropic systems of particles, the averaging

$$\langle \sigma_{ij}^{\alpha\beta}(i)\sigma_{ij}^{\delta\gamma}(j) \rangle_{\mathbf{r}_{ij}=\mathbf{r}} \quad (11)$$

should not depend on the direction of \mathbf{r} , while it can depend on $r = |\mathbf{r}|$. This is essentially what isotropicity means.

B. Transformation of correlation functions under rotations

Our goal in this section is to express the correlation functions between the stress tensor components in an arbitrary frame in terms of the correlation functions in the \mathbf{r}_{ij} frame introduced in the previous subsection.

If the direction from atom i to atom j forms angle θ_{ij} with the x axis of our reference coordinate frame, then the x axis of the directional coordinate frame also forms angle θ_{ij} with the x axis of our reference frame. Therefore, in order to find the stress tensor in our reference coordinate frame it is necessary to rotate the stress tensor in the representation associated with the directional coordinate frame on angle $-\theta_{ij}$. Thus, let us express the product $\sigma^{xy}(i)\sigma^{xy}(j)$ in the reference coordinate frame, which is rotated on the angle $-\theta_{ij}$ with respect to \mathbf{r}_{ij} frame in terms of stress tensor components in the \mathbf{r}_{ij} frame.

For this we should rotate, according to (5), (6), (7), the stress tensor components of atoms i and j in the \mathbf{r}_{ij} frame on the angle $-\theta_{ij}$ and then form the products of the stress tensor components in the rotated frame. From (7) we get:

$$\begin{aligned} \sigma_{\theta_{ij}}^{xy}(i)\sigma_{\theta_{ij}}^{xy}(j) &= (1/4)[\sigma_{ij}^{xx}(i) - \sigma_{ij}^{yy}(i)] \\ &\quad \times [\sigma_{ij}^{xx}(j) - \sigma_{ij}^{yy}(j)][\sin(2\theta_{ij})]^2 \\ &\quad + (1/4)[\sigma_{ij}^{xx}(i) - \sigma_{ij}^{yy}(i)]\sigma_{ij}^{xy}(j)[\sin(4\theta_{ij})] \\ &\quad + (1/4)\sigma_{ij}^{xy}(i)[\sigma_{ij}^{xx}(j) - \sigma_{ij}^{yy}(j)][\sin(4\theta_{ij})] \\ &\quad + \sigma_{ij}^{xy}(i)\sigma_{ij}^{xy}(j)[\cos(2\theta_{ij})]^2. \end{aligned} \quad (12)$$

The right-hand side of (12) can be expanded in terms containing the products of the stress tensor components in the \mathbf{r}_{ij} frame.

Let us now average (12) over the pairs of atoms i and j separated by $\mathbf{r}_{ij} = \mathbf{r}$ (this fixes the value of $\theta_{ij} = \theta$). For brevity and as an example let us consider a particular term, $\langle \sigma_{ij}^{xy}(i)\sigma_{ij}^{xy}(j)[\cos(2\theta_{ij})]^2 \rangle_{\mathbf{r}_{ij}=\mathbf{r}}$, that appears on the right-hand side of (12). In performing the averaging we get:

$$\langle \sigma_{ij}^{xy}(i)\sigma_{ij}^{xy}(j)[\cos(2\theta_{ij})]^2 \rangle_{\mathbf{r}_{ij}=\mathbf{r}} \quad (13)$$

$$= \langle \sigma_{ij}^{xy}(i)\sigma_{ij}^{xy}(j) \rangle_{\mathbf{r}_{ij}=\mathbf{r}} [\cos(2\theta)]^2. \quad (14)$$

In the transition from (13) to (14) the $[\cos(2\theta_{ij})]^2$ was taken out of the averaging since the averaging is performed for a fixed value of $\mathbf{r}_{ij} = \mathbf{r}$ and it also means that the averaging is performed for a fixed value $\theta_{ij} = \theta$. It follows from the previous section (Sec. IV A) that in isotropic medium $\langle \sigma_{ij}^{xy}(i)\sigma_{ij}^{xy}(j) \rangle_{\mathbf{r}_{ij}=\mathbf{r}}$ should not depend on the direction of \mathbf{r} , but can depend on $r = |\mathbf{r}|$.

Thus, in performing the averaging of the products of the stress tensor components in (12), as it was done in (13), (14), it is possible to average over all pairs of atoms separated by $\mathbf{r}_{ij} = \mathbf{r}$ irrespective of the direction of \mathbf{r} . It is only necessary to ensure that the values of the stress tensor components on the right-hand side of (12) are always calculated in the directional \mathbf{r}_{ij} frame corresponding to each pair of atoms i and j .

It follows from the above considerations that the value of the correlation function $\langle \sigma^{xy}(i)\sigma^{xy}(j) \rangle_{\mathbf{r}_{ij}=\mathbf{r}}$ at some r and θ can be expressed as a linear combination of the correlation functions between the atomic-level stress elements in the \mathbf{r}_{ij} frame multiplied on some functions of θ . Note that the dependence on θ in (12) appears in the result of rotation from the \mathbf{r}_{ij} frame into the frame in which \mathbf{r}_{ij} forms angle θ with the \hat{x} axis. Thus, in an isotropic medium the physical essence of the atomic-level stress correlations is contained in the correlation functions associated with the \mathbf{r}_{ij} frame. In an isotropic medium these correlation functions should depend only on distance.

C. Expressions for the selected stress correlation functions in terms of eigenvalues and eigenvectors in the \mathbf{r}_{ij} directional frame

It follows from the two previous sections (Secs. IV A, IV B) that in order to find correlation functions of the atomic-level stress components in any coordinate frame it is sufficient to know correlation functions in the directional \mathbf{r}_{ij} frame.

It is easy to express the correlation functions in the \mathbf{r}_{ij} frame in terms of eigenvalues and eigenvectors of the atomic-level stress matrices. Let us suppose that the first eigenvectors of the stress matrices of atoms i and j form angles ψ_{ij} and ψ_{ji} with the direction \mathbf{r}_{ij} , as shown in Fig. 2.

From (8), (9) it follows that the rotation invariant atomic-level pressure on atom i is

$$p_i \equiv (1/2)[\sigma_{ij}^{xx}(i) + \sigma_{ij}^{yy}(i)] = (1/2)(\lambda_i^1 + \lambda_i^2). \quad (15)$$

Correspondingly,

$$\langle p_i p_j \rangle_{\mathbf{r}_{ij}=\mathbf{r}} = (1/4)[(\lambda_i^1 + \lambda_i^2)(\lambda_j^1 + \lambda_j^2)]_{\mathbf{r}_{ij}=\mathbf{r}}. \quad (16)$$

It also follows from (8), (9), (10) that:

$$\langle p_i \sigma_{ij}^{xy}(j) \rangle_{\mathbf{r}_{ij}=\mathbf{r}} = (1/4)[(\lambda_i^1 + \lambda_i^2)(\lambda_j^1 - \lambda_j^2) \sin(2\psi_{ji})]_{\mathbf{r}_{ij}=\mathbf{r}}, \quad (17)$$

$$\begin{aligned} \langle p_i [\sigma_{ij}^{xx}(j) - \sigma_{ij}^{yy}(j)] \rangle_{\mathbf{r}_{ij}=\mathbf{r}} \\ = (1/2)[(\lambda_i^1 + \lambda_i^2)(\lambda_j^1 - \lambda_j^2) \cos(2\psi_{ji})]_{\mathbf{r}_{ij}=\mathbf{r}}, \end{aligned} \quad (18)$$

$$\begin{aligned} \langle \sigma_{ij}^{xy}(i)\sigma_{ij}^{xy}(j) \rangle_{\mathbf{r}_{ij}=\mathbf{r}} \\ = (1/4)[(\lambda_i^1 - \lambda_i^2)(\lambda_j^1 - \lambda_j^2) [\sin(2\psi_{ij}) \sin(2\psi_{ji})]]_{\mathbf{r}_{ij}=\mathbf{r}}, \end{aligned} \quad (19)$$

$$\begin{aligned} \langle \sigma_{ij}^{xy}(i) [\sigma_{ij}^{xx}(j) - \sigma_{ij}^{yy}(j)] \rangle_{\mathbf{r}_{ij}=\mathbf{r}} \\ = (1/2)[(\lambda_i^1 - \lambda_i^2)(\lambda_j^1 - \lambda_j^2) [\sin(2\psi_{ij}) \cos(2\psi_{ji})]]_{\mathbf{r}_{ij}=\mathbf{r}}, \end{aligned} \quad (20)$$

$$\begin{aligned} \langle [\sigma_{ij}^{xx}(i) - \sigma_{ij}^{yy}(i)] [\sigma_{ij}^{xx}(j) - \sigma_{ij}^{yy}(j)] \rangle_{\mathbf{r}_{ij}=\mathbf{r}} \\ = [(\lambda_i^1 - \lambda_i^2)(\lambda_j^1 - \lambda_j^2) [\cos(2\psi_{ij}) \cos(2\psi_{ji})]]_{\mathbf{r}_{ij}=\mathbf{r}}. \end{aligned} \quad (21)$$

Note that the right-hand sides of (15)–(21) depend on the invariant parameters of the atomic-level stress ellipses and on their rotation invariant orientations with respect to the direction of \mathbf{r}_{ij} . Thus, in finding how (16)–(21) depend on r in isotropic systems it is possible to average over all pairs separated by r irrespective of the orientation of \mathbf{r} . This is in agreement with the argument from Sec. IV A that states that correlation functions between the components of atomic-level stresses in the directional $\mathbf{r}_{ij} = \mathbf{r}$ frame should not depend on the direction of \mathbf{r} .

D. Stress correlation function $\langle \sigma^{xy}(i)\sigma^{xy}(j) \rangle_{\mathbf{r}_{ij}=\mathbf{r}}$ in the arbitrary reference frame expressed in terms of eigenvalues and eigenvectors

The correlation function $\langle \sigma^{xy}(i)\sigma^{xy}(j) \rangle_{\mathbf{r}_{ij}=\mathbf{r}}$ in any fixed reference frame depends on \mathbf{r} , i.e., on r and θ . Using expressions (12)–(14), (19)–(21) it is straightforward (although a bit tedious) to obtain the following expression:

$$\begin{aligned} \langle \sigma^{xy}(i)\sigma^{xy}(j) \rangle_{\mathbf{r}_{ij}=\mathbf{r}} \\ = (1/8)[F_1(r) - F_2(r) \cos(4\theta) + F_3(r) \sin(4\theta)], \end{aligned} \quad (22)$$

where

$$\begin{aligned} F_1(r) &\equiv \langle \mathcal{F}_1 \rangle_{\mathbf{r}_{ij}=\mathbf{r}} \\ &= [(\lambda_i^1 - \lambda_i^2)(\lambda_j^1 - \lambda_j^2) \cos(2\psi_{ij} - 2\psi_{ji})]_{\mathbf{r}_{ij}=\mathbf{r}}, \end{aligned} \quad (23)$$

$$\begin{aligned} F_2(r) &\equiv \langle \mathcal{F}_2 \rangle_{\mathbf{r}_{ij}=\mathbf{r}} \\ &= [(\lambda_i^1 - \lambda_i^2)(\lambda_j^1 - \lambda_j^2) \cos(2\psi_{ij} + 2\psi_{ji})]_{\mathbf{r}_{ij}=\mathbf{r}}, \end{aligned} \quad (24)$$

$$\begin{aligned} F_3(r) &\equiv \langle \mathcal{F}_3 \rangle_{\mathbf{r}_{ij}=\mathbf{r}} \\ &= [(\lambda_i^1 - \lambda_i^2)(\lambda_j^1 - \lambda_j^2) \sin(2\psi_{ij} + 2\psi_{ji})]_{\mathbf{r}_{ij}=\mathbf{r}}. \end{aligned} \quad (25)$$

Note that the dependence of (22) on θ originates from (12), i.e., from the rotation from the directional \mathbf{r}_{ij} frame into the coordinate frame that forms angle θ with the direction of

$r_{ij} = \mathbf{r} = [r \cos(\theta), r \sin(\theta)]$. Thus the dependence of (22) on θ merely reflects the rotational properties of the stress tensors. Also note that all physically meaningful information about correlations between the parameters of atomic-level stresses is contained in functions $F_1(r)$, $F_2(r)$, and $F_3(r)$.

In finding $F_1(r)$, $F_2(r)$, and $F_3(r)$ in isotropic medium the averaging can be performed over all pairs of atoms i and j separated by distance r irrespectively of the direction of \mathbf{r} .

Two alternative derivations of the formula (22) are presented in Appendixes A, B. In particular, Appendix A contains a very formal and general derivation of the above angular dependencies from the rotational properties of the stress tensors.

In order to understand the meaning of correlation function F_1 let us consider the contribution from some atoms i and j to this function. It follows from (23) that:

(i) If one of the ellipses is a circle, for example $\lambda_i^1 = \lambda_i^2$, then the contribution from this pair of atoms is zero. Thus, correlation function $F_1(r)$ contains contributions only from those pairs of atoms in which there are finite shear deformations of the environments of both atoms.

(ii) If the ellipses of atoms i and j have the same orientation with respect to the line connecting them then $\cos(2\psi_{ij} - 2\psi_{ji}) = 1$ and the contribution from this pair of ellipses is the maximum possible contribution from the pairs of ellipses with the same distortions.

(iii) If the ellipses of atoms i and j are orthogonal to each other, i.e., $\psi_{ij} = \psi_{ji} \pm \pi/2$ then $\cos(2\psi_{ij} - 2\psi_{ji}) = -1$ and the contribution from this pair is the minimum possible contribution.

(iv) If $\psi_{ij} = \psi_{ji} \pm \pi/4$ then the contribution is zero.

Note also the following. If the large axes of the ellipses of atoms i and j are aligned then these ellipses have the same orientation with respect to any line, not only the line that connects them. Thus, it is likely that rather simple organization of ellipses provides a maximum to the function F_1 . It is the organization when all ellipses have the same shear distortions and the same orientations. This observation might be of interest for understanding the nature of viscosity. It follows from the Green-Kubo expression that viscosity is determined by decay in time of the function $F_1(r)$, i.e., for calculations of viscosity it is necessary to consider stress of atom i at time zero and stress of atom j at time t [$F_2(r)$ does not contribute since integration over θ in (22) leads to zero].

In order to understand the meaning of correlation function $F_2(r)$ from (24) note the following:

(i) As in the case with $F_1(r)$, only pairs of atoms in which both atoms have shear distortions contribute.

(ii) The maximum contribution, for the given distortions, comes from the ellipses for which $\psi_{ij} = -\psi_{ji}$, i.e., from those ellipses whose orientations are mirror symmetric with respect to the line connecting them.

(iii) If the deviation from the mirror symmetry is $\pi/2$, i.e., $\psi_{ij} = -\psi_{ji} \pm \pi/2$ then the contribution is the minimum possible contribution.

(iv) If $\psi_{ij} = -\psi_{ji} \pm \pi/4$ then the contribution is zero.

Due to a mirror symmetry we must have $F_3(r) = 0$. This is because reflection with respect to the direction from i to j changes the signs of angles ψ_{ij} and ψ_{ji} , but does not change

the eigenvalues. In our simulations $F_3(r)$ averages to zero up to the noise level.

E. Stress correlation function $\langle p_i \sigma^{xy}(j) \rangle_{r_{ij}=r}$

From (7), (15), (17), (18), similarly to how it was done for $\langle \sigma^{xy}(i) \sigma^{xy}(j) \rangle_{r_{ij}=r}$, we get:

$$\langle p_i \sigma_j^{xy} \rangle_{r_{ij}=r} = (1/4)[F_4(r) \cos(2\theta) + F_5(r) \sin(2\theta)], \quad (26)$$

where

$$F_4(r) = \langle (\lambda_i^1 + \lambda_i^2)(\lambda_j^1 - \lambda_j^2) \sin(2\psi_{ji}) \rangle_{r_{ij}=r}, \quad (27)$$

$$F_5(r) = \langle (\lambda_i^1 + \lambda_i^2)(\lambda_j^1 - \lambda_j^2) \cos(2\psi_{ji}) \rangle_{r_{ij}=r}. \quad (28)$$

In finding $F_4(r)$ and $F_5(r)$ in isotropic medium the averaging can be performed over all pairs of atoms i and j separated by distance r irrespectively of the direction of \mathbf{r} .

Due to mirror symmetry, the function F_4 should average to zero (it does in simulations).

In order to understand the meaning of F_5 from (28) note the following:

(i) The larger is the pressure on atom i and the shear distortion of atom j , the larger is the contribution from this pair to F_5 .

(ii) If the ellipse of atom j is aligned with the direction from i to j then $\cos(2\psi_{ji}) = 1$ and there is the maximum possible contribution for the given ellipses' shapes.

(iii) If the ellipse of atom j is orthogonal to the direction from i to j then $\cos(2\psi_{ji}) = -1$ and there is the minimum possible contribution for the given ellipses' shapes.

(iv) If $\psi_{ji} = \pi/4$ then the contribution is zero.

F. Stress correlation function $\langle (\sigma_i^{xx} - \sigma_i^{yy}) \sigma_j^{xy} \rangle_{r_{ij}=r}$

From (5)–(7) and (19)–(21), similarly to how it was done for $\langle \sigma_i^{xy} \sigma_j^{xy} \rangle_{r_{ij}=r}$, we get:

$$\langle (\sigma_i^{xx} - \sigma_i^{yy}) \sigma_j^{xy} \rangle_{r_{ij}=r} = (1/2)F_1(r) \sin(4\theta) + F_6(r) \cos(4\theta), \quad (29)$$

where $F_1(r)$ is given by expression (23) and:

$$F_6(r) = \langle (\lambda_i^1 - \lambda_i^2)(\lambda_j^1 - \lambda_j^2) \cos(2\psi_{ij}) \sin(2\psi_{ji}) \rangle_{r_{ij}=r}. \quad (30)$$

In finding $F_6(r)$ in an isotropic medium the averaging can be performed over all pairs of atoms i and j separated by distance r irrespectively of the direction of \mathbf{r} .

The function F_6 should average to zero due to mirror symmetry with respect to the direction from i to j since under reflection $\cos(\psi_{ij})$ does not change sign, while $\sin(\psi_{ji})$ does. We verified this in our simulations.

G. Simpler correlation functions and normalization of the correlation functions

Correlation functions $F_{1,2,3,4,5,6}$ are somewhat complicated as they represent averages over three or four parameters. Before considering them it makes sense to consider simpler correlation functions, which represent averaged products of two parameters only. It is expected that stresses of particles that

are far away from each other are not correlated. This makes it reasonable to consider the following correlation functions:

$$G_{pp}(r) = (1/Z_+^2) \langle (\lambda_i^1 + \lambda_i^2)(\lambda_j^1 + \lambda_j^2) \rangle_{r_{ij}=r} - 1, \quad (31)$$

$$G_{mm}(r) = (1/Z_-^2) \langle (\lambda_i^1 - \lambda_i^2)(\lambda_j^1 - \lambda_j^2) \rangle_{r_{ij}=r} - 1, \quad (32)$$

$$G_{mp}(r) = (1/Z_+Z_-) \langle (\lambda_i^1 - \lambda_i^2)(\lambda_j^1 + \lambda_j^2) \rangle_{r_{ij}=r} - 1, \quad (33)$$

$$C_{2\pm}(r) = \langle \cos(2\psi_{ij} \pm 2\psi_{ji}) \rangle_{r_{ij}=r}, \quad (34)$$

where $Z_{\pm} = \langle \lambda_i^1 \pm \lambda_i^2 \rangle$.

Functions $G_{pp}(r)$, $G_{mm}(r)$, and $G_{mp}(r)$ describe correlations between the eigenvalues (or eigenstresses) of the stress matrices of atoms i and j without taking into account the orientations of the eigenvectors. Note that since $p_i = (1/2)(\lambda_i^1 + \lambda_i^2)$ the function $G_{pp}(r)$ from (31) is directly related to the pressure-pressure correlation function between atoms i and j . It follows from Appendix A and formula (2) that the function G_{mm} represents correlations between the total amounts of shear on atoms i and j . Finally, $G_{mp}(r)$ describes correlations between the total shear on atom i and the total pressure on atom j . Functions $C_{2\pm}(r)$ from (34) describe correlations in the orientations of the eigenvectors of the stress matrices of atoms i and j without taking into account the magnitudes of the eigenvalues.

It is also reasonable to introduce normalized versions of the correlation functions $F_{1,2,3,4,5,6}$:

$$\tilde{F}_{1,2,3,6}(r_{ij}) \equiv F_{1,2,3,6}/Z_-^2, \quad (35)$$

$$\tilde{F}_{4,5} \equiv F_{4,5}/(Z_+Z_-). \quad (36)$$

V. ANALOGY WITH ESHELBY'S INCLUSION PROBLEM

The ability of liquids to support shear stresses on the timescales comparable to Einstein's vibrational periods (and larger) is well known. An early example of the relevant considerations is represented by the Maxwell's viscoelastic model [19]. Recent investigations of the propagation of shear stresses in glasses and liquids demonstrated, in particular, their ability to support anisotropic shear stresses. Thus, it has been shown that ellipselike deformations of small (≈ 10 – 20 particles) circular regions in glasses and liquids generate atomic displacements and shear stress fields with angular dependencies similar to those produced by Eshelby's inclusion in a continuous medium [20–28]. For this reason we discuss in this section, from the perspective of Eshelby's inclusion problem [18,29–32], the stress correlation function, which is analogous to the atomic-level stress correlation function $\langle \sigma_i^{xy} \sigma_j^{xy} \rangle_{r_{ij}=r}$. In particular, we argue that the angular dependence of the $\langle \sigma_i^{xy} \sigma_j^{xy} \rangle_{r_{ij}=r}$ stress correlation function obtained in Ref. [14] is related to the rotational properties of the stress tensors and not to the anisotropy of the stress field associated with Eshelby's solution, as it is suggested in Ref. [14].

In drawing the analogy with Eshelby's inclusion problem, we assume that every atom i with its nearest-neighbor shell is analogous to Eshelby's inclusion, I , which generates a stress field in the matrix at point J (atomic stress on atom j). There are two points that we need from Eshelby's solution.

(i) The final constrained strain and stress fields are the same everywhere in the inclusion. These fields, of course, depend on the unconstrained strain initially applied to the inclusion.

(ii) If the unconstrained strain applied to the inclusion is known then the final constrained strain and stress fields in the inclusion and in the matrix can be found. Further, it is assumed that there is a one-to-one correspondence between the constrained stress fields in the inclusion and in the matrix. See also Appendix C.

We are interested in the correlation functions between the inclusion, I , and some point, J , in the matrix. Similarly to how it was done for the atomic-level stresses, we can associate with I the stress ellipse whose parameters, $(\lambda_I^1, \lambda_I^2)$, and whose orientation, ψ_{IJ} , with respect to the \mathbf{r}_{IJ} are known. The fact that the constrained stress field is the same everywhere in the inclusion serves well for this purpose.

Since the inclusion's stress ellipse is known, the stress field (tensor) at any point J in the matrix can be found. Therefore it is possible to associate with point J its own stress ellipse with parameters $(\lambda_J^1, \lambda_J^2)$ and the orientation ψ_{JI} with respect to the \mathbf{r}_{IJ} . Thus, λ_J^1 , λ_J^2 , and ψ_{JI} are the functions of λ_I^1 , λ_I^2 , ψ_{IJ} , and r_{IJ} :

$$\lambda_J^1 = \lambda_J^1(\lambda_I^1, \lambda_I^2, \psi_{IJ}, r_{IJ}), \quad (37)$$

$$\lambda_J^2 = \lambda_J^2(\lambda_I^1, \lambda_I^2, \psi_{IJ}, r_{IJ}), \quad (38)$$

$$\psi_{JI} = \psi_{JI}(\lambda_I^1, \lambda_I^2, \psi_{IJ}, r_{IJ}). \quad (39)$$

In (37)–(39) angles ψ_{IJ} and ψ_{JI} are the angles between the larger ellipses' axes and the direction \mathbf{r}_{IJ} . Note that in isotropic elastic medium λ_I^1 , λ_I^2 , and ψ_{JI} should not depend on the direction of \mathbf{r}_{IJ} (the direction of the inclusion's deformation with respect to \mathbf{r}_{IJ} is taken into account by the angle ψ_{IJ}). Note also that the properties of Eshelby's solution are embedded into (37)–(39). These functions, in our view, represent the essence of Eshelby's solution. In Appendix C a particular case of the inclusion's shear transformation is discussed and functions (37)–(39) for this case are derived.

At this point it becomes apparent that considerations of the correlations for Eshelby's inclusion problem are quite similar to the considerations that were already done for the atomic-level stresses. Thus, the expressions for the stress correlation functions between the inclusion and the matrix can be derived in the same way as the expressions (22)–(26), (29) for the atomic-level stress correlation functions. For example, for the product $\sigma^{xy}(I)\sigma^{xy}(J)$ we get (see Ref. [33] for a brief reminder on how the formula is derived):

$$\sigma^{xy}(I)\sigma^{xy}(J) = (1/8)[\mathcal{F}_1^e - \mathcal{F}_2^e \cos(4\theta_{IJ}) + \mathcal{F}_3^e \sin(4\theta_{IJ})], \quad (40)$$

where θ_{IJ} is the angle between \mathbf{r}_{IJ} and the x axis of the reference Cartesian coordinate frame and

$$\mathcal{F}_1^e \equiv (\lambda_I^1 - \lambda_I^2)(\lambda_J^1 - \lambda_J^2) \cos(2\psi_{IJ} - 2\psi_{JI}), \quad (41)$$

$$\mathcal{F}_2^e \equiv (\lambda_I^1 - \lambda_I^2)(\lambda_J^1 - \lambda_J^2) \cos(2\psi_{IJ} + 2\psi_{JI}), \quad (42)$$

$$\mathcal{F}_3^e \equiv (\lambda_I^1 - \lambda_I^2)(\lambda_J^1 - \lambda_J^2) \sin(2\psi_{IJ} + 2\psi_{JI}). \quad (43)$$

The upper index e in the formulas above originates from the word “elastic”. Note again that λ_j^1 , λ_j^2 , and ψ_{IJ} in (41)–(43) are the functions of λ_j^1 , λ_j^2 , ψ_{IJ} , and r_{IJ} . Also note that \mathcal{F}_1^e , \mathcal{F}_2^e , \mathcal{F}_3^e do not depend on θ_{IJ} . Thus in (41)–(43)

$$\mathcal{F}_n^e = \mathcal{F}_n^e(\lambda_j^1, \lambda_j^2, \psi_{IJ}, r_{IJ}), \text{ where } n = 1, 2, 3, \quad (44)$$

i.e., functions \mathcal{F}_1^e , \mathcal{F}_2^e , and \mathcal{F}_3^e are determined by how the stress field in the inclusion determines the stress field at point J .

Now we address the connection between the functions \mathcal{F}_1^e , \mathcal{F}_2^e , and \mathcal{F}_3^e from (41)–(43) and the functions F_1 , F_2 , and F_3 from (23)–(25). The functions \mathcal{F}_1^e , \mathcal{F}_2^e , and \mathcal{F}_3^e are written for a particular set of values λ_j^1 , λ_j^2 , ψ_{IJ} , and r_{IJ} . In order to draw a parallel with the atomic-level stress correlation functions in liquids it is necessary to average the functions \mathcal{F}_1^e , \mathcal{F}_2^e , and \mathcal{F}_3^e over the possible values of λ_j^1 , λ_j^2 , and ψ_{IJ} which can be associated with the parameters of the inclusion’s stress ellipse. Thus:

$$F_n^e(r_{IJ}) = \langle \mathcal{F}_n^e(\lambda_j^1, \lambda_j^2, \psi_{IJ}, r_{IJ}) \rangle_{\lambda_j^1, \lambda_j^2, \psi_{IJ}}, \quad (45)$$

where $n = 1, 2, 3$. In (45) it is presumed that every set of parameters at I deterministically leads to certain parameters at J via Eshelby’s solution. In (45) there is no averaging over the distance (scalar) r_{IJ} . Correspondingly functions F_n^e depend only on $r_{IJ} \equiv r$.

In liquids there is no deterministic relationship between the parameters and orientations of the atomic-level stress ellipses of atoms i and j . In liquids there is only a probabilistic relationship. Thus in calculations of $F_1(r)$, $F_2(r)$, and $F_3(r)$ in liquids (23)–(25) the averaging goes not only over λ_i^1 , λ_i^2 , ψ_{ij} , but also over λ_j^1 , λ_j^2 , ψ_{ji} . Implicitly in calculations of (23)–(25) there is also the averaging over the directions of \mathbf{r}_{ij} for a fixed value of r_{ij} . In the case of Eshelby’s inclusion it is assumed that the undistorted inclusion and the matrix are isotropic. Thus, there is no need to average (45) over the directions of \mathbf{r}_{IJ} .

Note that if $\langle \sigma_I^{xy} \sigma_J^{xy} \rangle$ were calculated from (B2) in a particular reference frame, by averaging over the possible distortions of the inclusion, it still would depend on θ_{IJ} . This dependence, however, does not represent the angular-dependent Eshelby’s stress field. Instead the dependence on θ_{IJ} in (B2) reflects the rotational properties of the stress tensors. The angular dependencies observed in Ref. [14] correspond to the dependence of $\langle \sigma^{xy}(I) \sigma^{xy}(J) \rangle$ on θ_{IJ} in (B2). This is not the angular dependence of Eshelby’s field. The angular dependence of Eshelby’s stress field is embedded in how λ_j^1 , λ_j^2 , ψ_{IJ} depend on λ_j^1 , λ_j^2 , ψ_{IJ} , and r_{IJ} .

Finally, note that the dependence of (B2) on θ_{IJ} does not depend on the type of the inclusion’s distortion or on the type of the dependence between the inclusion’s distortion and the stress field in the matrix. The same angular dependence would occur for any type of inclusion’s distortion and for any type of connection between the stress fields in the inclusion and in the matrix. All information about the nature of the inclusion’s distortion and the connection between the stress fields in the inclusion and in the matrix is contained in functions $\langle \mathcal{F}_n^e(\lambda_j^1, \lambda_j^2, \psi_{IJ}, r_{IJ}) \rangle$.

VI. RESULTS OF MD SIMULATION

A. Stress correlation functions

In our molecular dynamics (MD) simulations we considered the same 2D one-component system of particles that has been studied in Ref. [14]. Particles interact through Yukawa potential, $U(r) = U_o(\frac{\sigma}{r}) \exp[-\lambda(\frac{\sigma-r}{\sigma})]$, where in Lennard-Jones units $U_o = 1$, $\sigma = 1$, $\lambda = 8$. The mass of every particle is also one, $m = 1$. MD simulations were performed using the LAMMPS MD program [34,35]. We studied the systems of two sizes. In the small system the number of particles was $N = 2500$, while the dimensions of the rectangular periodic box were $L_x = 50.1021$, $L_y = 43.3897$. Our small system has the same size as the system studied in Ref. [14]. Another (large) system that we studied contained $N = 22500$ particles, i.e., nine times more than the small system. The dimensions of the large system were $L_x = 150.306$, $L_y = 130.169$. The particles’ number densities in the small and large systems are the same $\rho_o = 1.15$. We performed simulations in NVT and NVE ensembles.

In all cases the systems were prepared by melting triangular lattice at reduced temperature $T = 5$. After the equilibration at $T = 5$ the temperature was reduced in several steps that followed by equilibration at every temperature (in NVT ensemble) or at every value of fixed total energy (in NVE ensemble). The temperature in NVT ensemble was introduced via Nosé-Hoover thermostat. The damping parameter corresponded to 100 MD steps and also to 0.1 of the time unit.

In our simulations, we reproduced the dependence of potential energy on temperature presented in Fig. 1 of Ref. [14].

Atomic configurations for calculations of the correlation functions related to the eigenvalues and eigenvectors of atomic-level stresses were collected on the small system in the NVE ensemble at total energies, which corresponded to the following temperatures: $T(3) = 3.06 \pm 0.04$, $T(2) = 1.97 \pm 0.03$, $T(1.4) = 1.43 \pm 0.02$, $T(1) = 0.99 \pm 0.02$. The averaging was done over 1000 configurations at every temperature. For the temperature $T = 1$ the time interval between the two consecutive configurations was 10^4 MD steps. Each MD step corresponded to 0.001 of the time unit. During these 10^4 MD steps the mean-square atomic displacement reaches $\sim 1.38\sigma$.

Different correlation functions per pair of particles are shown in Figs. 3, 4, 5. The dependencies of the functions \tilde{F}_1 , \tilde{F}_2 and \tilde{F}_3 , i.e., all the nonzero ones, on distance are shown in Fig. 5. At $T = 1$ we have $Z_- = \langle \lambda_i^1 - \lambda_i^2 \rangle \approx 10.82$ and $Z_+ = \langle \lambda_i^1 + \lambda_i^2 \rangle \approx 40.48$.

Figures 3, 4, 5 demonstrate that there are r_{ij} -dependent correlations between the parameters of the atomic-level stress ellipses and in their orientations. These correlations gradually decrease with increase of r_{ij} . It is clear that functions $G_{mm}(r)$ in Fig. 3(b) and $\langle \cos(2\psi_{ij} - 2\psi_{ji}) \rangle$ in Fig. 4(a) exhibit more pronounced changes than does PDF [Fig. 3(a)] on decrease of temperature. It is also clear that the first peaks in \tilde{F}_1 and \tilde{F}_2 [Figs. 5(a), 5(b)] also demonstrate more pronounced changes on decrease of temperature than does PDF. However, it is also more difficult to interpret these changes. Yet, developing features in $\langle \cos(2\psi_{ij} - 2\psi_{ji}) \rangle$ suggest that some ordering happens in the mutual orientations of the ellipses associated with the atoms separated by the distance corresponding the

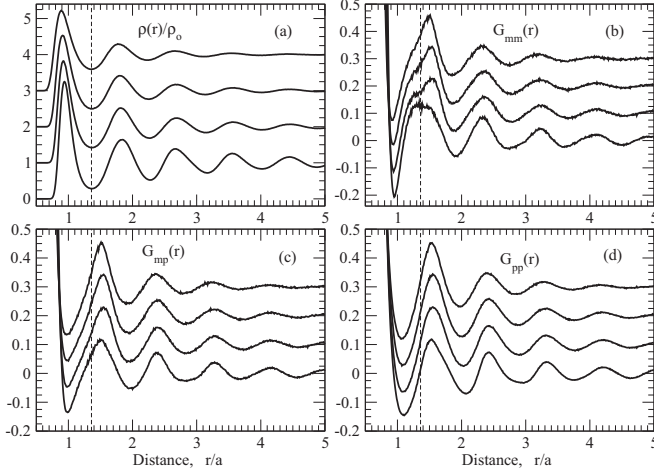


FIG. 3. In every panel the curves from the top to the bottom correspond to temperatures: $T = 3$, $T = 2$, $T = 1.4$, and $T = 1$. The curves were shifted vertically for the clarity of the presentation. (a) Pair density function. (b) G_{mm} correlation function from (32). (c) G_{mp} correlation function from (33). (d) G_{pp} correlation function from (31).

first minimum in the PDF. There also appears to be a certain similarity in the behaviors of $\langle \cos(2\psi_{ij} - 2\psi_{ji}) \rangle$ and \tilde{F}_1 . This similarity suggests that changes in \tilde{F}_1 are caused by changes in $\langle \cos(2\psi_{ij} - 2\psi_{ji}) \rangle$. See expression (23) for F_1 . Thus, changes

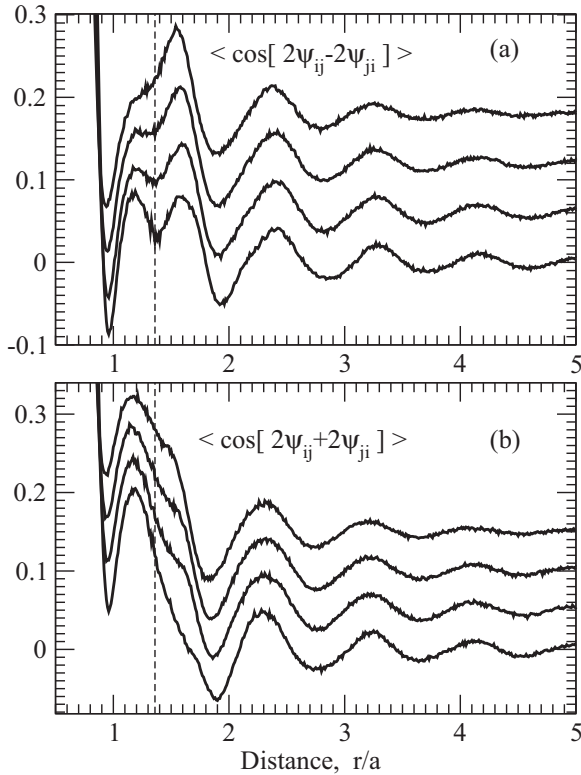


FIG. 4. Evolutions with temperature of the functions $\langle \cos(2\psi_{ij} - 2\psi_{ji}) \rangle$ and $\langle \cos(2\psi_{ij} + 2\psi_{ji}) \rangle$. In both panels the curves from the top to the bottom correspond to temperatures $T = 3$, $T = 2$, $T = 1.4$, and $T = 1$. The curves were shifted vertically for the clarity of the presentation.

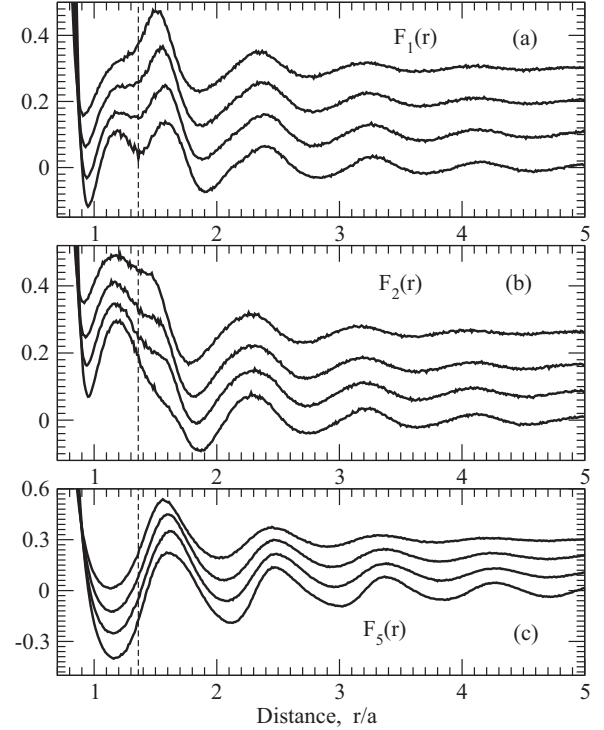


FIG. 5. Evolutions with temperature of the normalized functions \tilde{F}_1 from (23), (35), \tilde{F}_2 from (24), (35), and \tilde{F}_5 from (28), (36). In every panel the curves from the top to the bottom correspond to temperatures $T = 3$, $T = 2$, $T = 1.4$, and $T = 1$. The curves were shifted vertically for the clarity of the presentation.

in \tilde{F}_1 are likely to be caused not by changes in the eigenvalues of the stress ellipses, but by changes in the mutual orientations of the ellipses. However, also note that there are changes in $G_{mm}(r)$ in Fig. 3(b).

Figure 6 shows how the function $\tilde{F}_1(r)\rho(r)$ changes with temperature. It follows from the figure that as temperature is reduced there develops a pronounced minimum at the position of the first minimum, r_{min}^1 , of $\rho(r)$. Thus, changes in $\tilde{F}_1(r)$

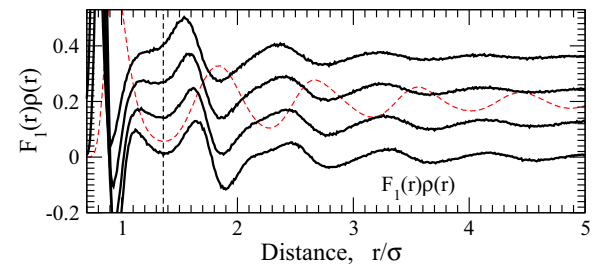


FIG. 6. Evolution with temperature of the function $\tilde{F}_1(r)\rho(r)$. The curves from the top to the bottom correspond to temperatures $T = 3$, $T = 2$, $T = 1.4$, and $T = 1$. The curves were shifted vertically for clarity of presentation. The dashed curve shows the scaled $\rho(r)$ at $T = 1.0$. As temperature decreases the first minimum in $\rho(r)$ becomes deeper. This deepening overlaps with the development of the minimum in $\tilde{F}_1(r)$. Thus, the development of the feature in $\tilde{F}_1(r)$ at the position of the first minimum of $\rho(r)$ is also well pronounced in $\tilde{F}_1(r)\rho(r)$.

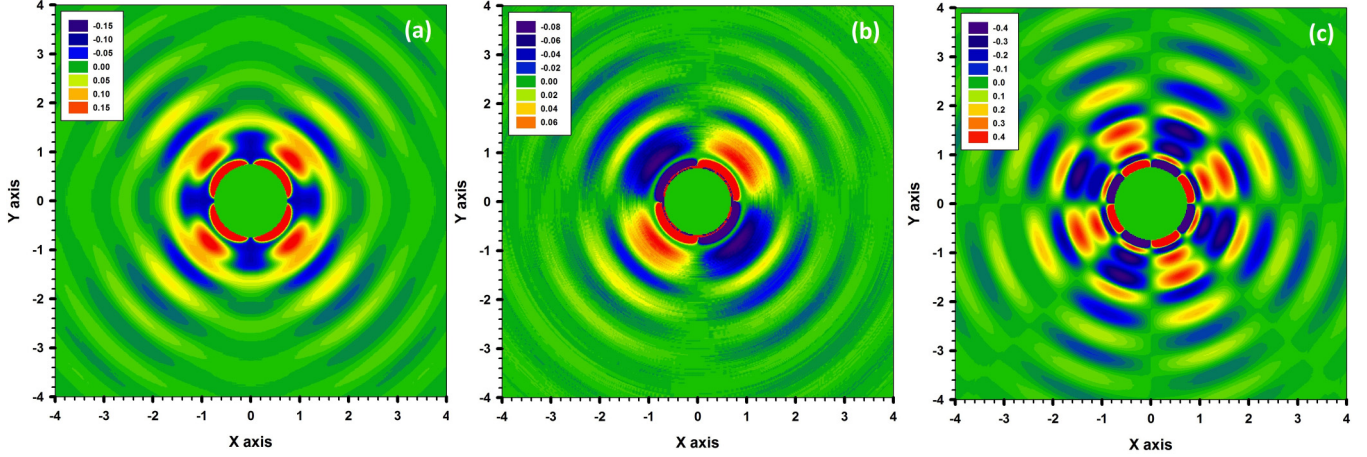


FIG. 7. (a) Atomic level stress correlation function $\langle \sigma_i^{xy} \sigma_j^{xy} \rangle$. See formula (22). The functions F_1 and F_2 in $\langle \sigma_i^{xy} \sigma_j^{xy} \rangle$ were normalized according to (35). The function F_3 is zero, besides the noise. It is clear that (a) of this figure is very similar to (a) of Fig. 5 in Ref. [14]. (b) Atomic level stress correlation function $\langle p_i \sigma_j^{xy} \rangle$. See formula (26). The function F_4 in $\langle p_i \sigma_j^{xy} \rangle$ averages to zero. Thus only function F_5 is left. In order to produce this figure we had to subtract the average pressure from the diagonal components of the atomic-level stress tensor. Effectively this means that the value of λ averaged over λ^1 and λ^2 of all atoms, i.e., λ^{ave} , was subtracted from the values of λ^1 and λ^2 of every atom. Because of this subtraction we can not use normalization (33) since $\langle \lambda^1 + \lambda^2 - 2\lambda^{\text{ave}} \rangle$ averages to zero. Thus we used normalization (32) instead. We also scaled intensity on the z -axis by a factor of 4. It is obvious that (b) of this figure is very similar to the (b) of Fig. 5 in Ref. [14]. (c) Atomic level stress correlation function $\langle (\sigma_i^{xx} - \sigma_i^{yy}) \sigma_j^{xy} \rangle$. See Eq. (29). Only function F_1 in $\langle (\sigma_i^{xx} - \sigma_i^{yy}) \sigma_j^{xy} \rangle$ is nonzero. We scaled the function by a factor of 10 along the z -intensity axis. It is clear that correlation function in (c) is rather similar to the correlation function in (c) of Fig. 5 in Ref. [14]. There is a difference with the two circles at $(r/a) \lesssim 1$. It is possible that authors of Ref. [14] did not look into such small distances and thus in their figure these circles fall into the central green region. Besides this difference their figures and ours look rather similar.

are also well observable in $\tilde{F}_1(r)\rho(r)$ despite the fact that the number of atomic pairs separated by r_{\min}^1 is relatively small.

The curves in Fig. 5 can be converted into the 2D intensity plots equivalent to those presented in Ref. [14] using formulas (22), (26), (29). Thus, if we want to find the stress correlation function at a point with coordinates (x, y) we define $r = \sqrt{x^2 + y^2}$ and $\theta = \arctan(y/x)$. Using these values in (22), (26), (29) the stress field at (x, y) can be found. This conversion applies because for particles i and j with coordinates (x_i, y_i) and (x_j, y_j) the values of r_{ij} and θ_{ij} that go into the formulas (22), (26), (29) are $r_{ij} = \sqrt{(x_j - x_i)^2 + (y_j - y_i)^2}$ and $\theta_{ij} = \arctan[(y_j - y_i)/(x_j - x_i)]$. However, in making the 2D stress correlation function plots it is assumed that the particle i is at the origin.

The results of the conversion described above for $T = 1$ are presented in Fig. 7. It is obvious that the 2D plots in Fig. 7 are very similar to those shown in Fig. 5 of Ref. [14]. Note that the 2D plots presented in Fig. 7 were obtained from only three functions, i.e., $F_1(r)$, $F_2(r)$, and $F_5(r)$, which depend only on r . This proves that the dependencies on θ presented in the 2D plots in Ref. [14] follow from the tensorial rotational properties.

B. Is the studied system in a hexatic phase at $T = 0.95$?

Role of size effects

Finally, we comment on the following statement made in Ref. [14]. It is stated there that at $T = 1$ the system is in a true liquid state, while at $T = 0.95$ the system is in a hexatic state.

We would like to emphasize that our goal in this section is not to make a definite conclusion about the state of the system at $T = 0.95$ (is it hexatic or not?). Instead, our goal

is to demonstrate that in considerations relevant to the hexatic phase and the bond-order correlation function in the discussed system it is necessary to consider the size effects. Thus, the data presented in this section give us an insight into the magnitude of the size effects in our system. They also address the length scales of the bond-order correlation function.

In order to make a distinction between the true liquid and hexatic states, it was assumed in Ref. [14] that in a true liquid state the bond-order correlation function decays exponentially with increase of distance, while in the hexatic state the bond-order correlation function decays algebraically. The following definition of the bond-order correlation function, $g_6(r)$, was used in Ref. [14]:

$$g_6(r) \equiv \langle \Psi_i^*(r) \Psi_j(0) \rangle, \quad \text{where} \quad \Psi_i(r) \equiv \frac{1}{N_k} \sum_{j=1}^{N_k} e^{i6\theta_{ik}}. \quad (46)$$

The summation over j in (46) goes over the neighbors of atom i . Two atoms are defined as neighbors if their separation distance is smaller than the position of the first minimum in the pair density function, i.e., $(r_{ij}/a) \leq 1.36\sigma$. The averaging in (46) goes over the pairs of atoms separated by distance r . In a perfect triangular lattice $g_6(r)$ is equal to 1 at distances that correspond to the lattice spacings and it is 0 otherwise.

We calculated how the bond-order correlation function (46) depends on distance in systems of two sizes. In the small system containing $N = 2500$ particles $(L_y/2) = 21.7$. In the large system $N = 22\,500$ and $(L_y/2) = 65.1$. The small system was used in Ref. [14]. The results are presented in Figs. 8, 9.

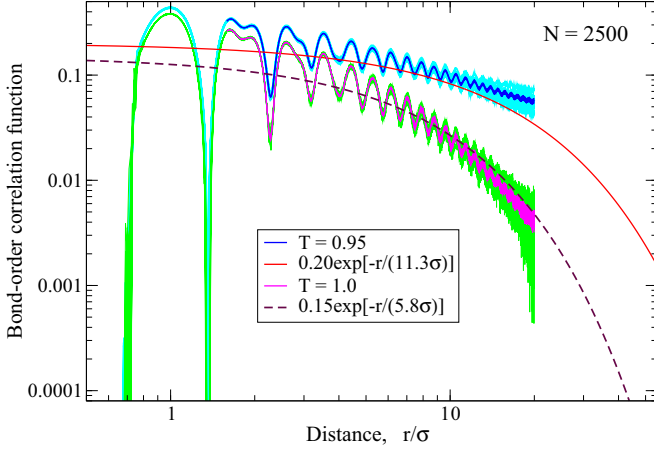


FIG. 8. Bond-order correlation functions for the system of $N = 2500$ particles. The upper set of curves is related to $T = 0.95$, while the lower set of curves is related to $T = 1.0$. There are 10 cyan curves in the upper set curves. Every cyan curve is the average over 1000 independent configurations. The mean-square displacement between the consecutive configurations was larger than the interatomic distance. There are also three blue curves in the upper set of curves. These blue curves represent the mean over the 10 cyan curves and the average \pm the error of the mean. Finally, in the top set of curves there is a smooth (red) curve that represents the fitting exponential function obtained on the large system of particles. There are 10 green curves in the bottom set of curves ($T = 1.0$). Every green curve represents the average over 100 independent configurations. In the lower set there are also three magenta curves which represent the average over the green curves and the average \pm the error of the mean. The dashed (maroon) curve in the lower set shows the exponential fit obtained from the larger system of $N = 22500$.

It follows from Figs. 8, 9 that on decrease of temperature sixfold rotational symmetry undoubtedly develops in the systems. The comparison of Fig. 8 with Fig. 9 suggests that at $T = 0.95$ the small system exhibits observable size effects. Note that at $T = 0.95$ the decay length is larger than $(1/2)$ of $(L_y/2)$ in the small system. Thus, in the small system the bond-order correlation function does not completely decay on the half length of the simulation box. It also follows from the data obtained on the large system that the exponential fit to the data is better than any algebraic fit can be at both temperatures. Thus, in our view, it follows from Figs. 8, 9 that it is impossible to make a qualitative distinction between the liquid states at $T = 1$ and $T = 0.95$ on the basis of the obtained data.

The observation of the hexatic phase was reported in Ref. [14] on the basis of the algebraic decay of the bond-order correlation function at $T = 0.95$ in the small system. The comparison of our data from the small and large systems demonstrates that algebraic decay of the bond-order correlation function in the small system at $T = 0.95$ is likely to originate from the size effects and thus it can not be reliably attributed to the formation of the hexatic phase.

VII. CONCLUSION

It was demonstrated that it is possible to study liquid (and glass) structures through considerations of correlations

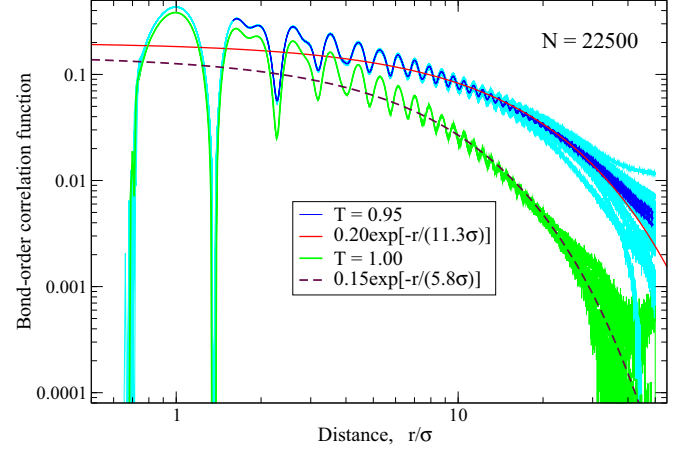


FIG. 9. Bond-order correlation functions for the system of $N = 22500$ particles. The upper and lower sets of curves are related to the temperatures $T = 0.95$ and $T = 1.0$ correspondingly. There are 21 cyan curves in the upper set of curves. Every cyan curve is the average over 100 independent configurations. The mean-square displacement between the consecutive configurations was larger than the interatomic distance. There are also three blue curves in the top set of curves. These blue curves represent the mean over the 21 cyan curves and the mean \pm the error of the mean curves. In the top set there is also smooth (red) curve that represents the exponential fit to the average blue curve. It is possible that at distances larger than $r/\sigma = 30$ at $T = 0.95$ there again appear size effects. There are three green curves in the bottom set of curves. Every green curve is the average over 200 independent configurations. The smooth dashed (maroon) curve represents exponential fit to the data.

between the eigenvalues and eigenvectors of the atomic-level stress tensors of different atoms. It was shown that on decrease of temperature some of the studied correlation functions exhibit pronounced changes in the range of distances that corresponds to the first minimum of the pair density function. These changes could not be guessed from the behavior of the pair density function. Thus, the suggested method provides additional information and it is of interest to investigate evolution of stress correlations with this method in model supercooled liquids on decrease of temperature.

We also demonstrated that interpretations of the angular dependencies of the stress correlation functions reported in Ref. [14] are essentially incorrect. In particular, the authors of Ref. [14] associate the angular dependencies observed in the stress correlation functions with the angular dependencies of Eshelby's stress field. We demonstrated that anisotropic stress fields observed in Ref. [14] originate from the rotational properties of the stress tensors. We also had shown that information that is really related to the anisotropic Eshelby's stress fields is embedded into the isotropic stress correlation functions $F_1(r)$, $F_2(r)$, and $F_5(r)$, which we studied in this work.

From a purely pragmatic perspective we have shown that eight 2D panels of the stress correlation functions presented in Ref. [14] can be reproduced using only three correlation functions, which depend only on r , i.e., from the functions $F_1(r)$, $F_2(r)$, and $F_5(r)$ [36]. This clearly advances understanding of the plots of the stress correlation functions presented

in Ref. [14]. It also follows from our results that instead of studying distance dependence of the integrals of the 2D stress correlation functions over some angles, as it has been done in Ref. [14], it is more reasonable to study how functions $F_1(r)$, $F_2(r)$, and $F_3(r)$ depend on distance. We also demonstrated that because of size effects the distinction made in Ref. [14] between the normal liquid and hexatic states is invalid.

APPENDIX A: ALTERNATIVE DERIVATION OF $\langle \sigma_i^{xy} \sigma_j^{xy} \rangle$ STRUCTURE

In two dimensions in a particular reference coordinate frame numerical representation of the atomic-level stress tensor $\hat{\sigma}$ is a 2×2 matrix. This matrix is real and symmetric (i.e., $\sigma^{yx} = \sigma^{xy}$), thus it can be diagonalized. We can work directly with its components $\sigma^{\alpha\beta}$; or with corresponding pressure p and two shear components, s_1 and s_2 ; or with real eigenvalues $\lambda_{1,2}$ and the orientation of two orthogonal eigenvectors:

$$\hat{\sigma} = \begin{bmatrix} \sigma^{xx} & \sigma^{xy} \\ \sigma^{xy} & \sigma^{yy} \end{bmatrix} = \begin{bmatrix} p + s_1 & s_2 \\ s_2 & p - s_1 \end{bmatrix} \\ = \hat{R}(\varphi) \begin{bmatrix} \lambda_1 & 0 \\ 0 & \lambda_2 \end{bmatrix} [\hat{R}(\varphi)]^T.$$

Here $\hat{R}(\varphi)$ is the 2×2 matrix of rotation in positive (or counterclockwise) direction by angle φ :

$$\hat{R}(\varphi) = \begin{bmatrix} \cos \varphi & -\sin \varphi \\ \sin \varphi & \cos \varphi \end{bmatrix}.$$

Pressure and shear components are expressed through $\sigma^{\alpha\beta}$ components as

$$p = \frac{1}{2}(\sigma^{xx} + \sigma^{yy}), \quad s_1 = \frac{1}{2}(\sigma^{xx} - \sigma^{yy}), \quad s_2 = \sigma^{xy}.$$

It will be convenient for us to combine the shear components into a single complex number $s = s_1 + i s_2$. The total amount of shear is given by its absolute value:

$$|s| = \sqrt{s_1^2 + s_2^2} = \sqrt{\frac{1}{4}(\sigma^{xx} - \sigma^{yy})^2 + (\sigma^{xy})^2},$$

while the argument of s is related to the shear's direction.

Consider three reference frames (x, y) , (x', y') , and (x'', y'') , with (x', y') being obtained from (x, y) by a rotation in negative (clockwise) direction by angle α , while (x, y) and (x'', y'') are mirror reflections of each other with respect to x axis. We will write down the quantities in (x', y') and (x'', y'') frames with prime and double prime symbols, respectively. The transformation properties of the stress tensor (5)–(7) result in

$$s' = s \exp(2i \alpha), \quad s'' = s^*,$$

where $*$ denotes complex conjugation.

Since $\sigma^{xy} = (s - s^*)/2i$, we have:

$$\langle \sigma_i^{xy} \sigma_j^{xy} \rangle = \frac{1}{16} [F_1(r, \theta) + F_1^*(r, \theta) - F_2(r, \theta) - F_2^*(r, \theta)], \quad (\text{A1})$$

where

$$F_1(r, \theta) = 4\langle s_i s_j^* \rangle, \quad F_2(r, \theta) = 4\langle s_i s_j \rangle; \quad (\text{A2})$$

All the averages $\langle \cdot \rangle$ are taken over the pairs of atoms i and j with $r_{ij} = r$ and $\theta_{ij} = \theta$.

By checking how s and the angle θ are transformed by rotations [$s' = s \exp(2i \alpha)$ and $\theta' = \theta + \alpha$] we get

$$F_1(r, \theta + \alpha) = F_1(r, \theta), \\ F_2(r, \theta + \alpha) = F_2(r, \theta) \exp(4i \alpha). \quad (\text{A3})$$

The function $F_1(r, \theta)$ does not depend on the angle θ at all. By considering $F_1^*(r, \theta)$ we exchange the roles of atoms i and j , which is equivalent to the change $\theta \rightarrow \theta + \pi$. Thus $F_1^*(r, \theta) = F_1(r, \theta + \pi) = F_1(r, \theta)$, i.e., we get $F_1 = F_1^*$. All this means that $F_1(r, \theta) = F_1(r)$ is a real function of a single parameter r .

If we put $\theta = 0$ in (A3), we get $F_2(r, \alpha) = F_2(r, 0) \exp(4i \alpha)$. Mirror reflection ($s'' = s^*$ and $\theta'' = -\theta$) leads to $F_2(r, -\theta) = F_2^*(r, \theta)$. In particular, $F_2(r) = F_2(r, 0) = F_2^*(r, 0)$ is also a real function of just the distance between the atoms r . Also, $F_2(r, \theta) = F_2(r) \exp(4i \theta)$.

Putting these results for F_1 and F_2 into the expression (A1) we finally get

$$\langle \sigma_i^{xy} \sigma_j^{xy} \rangle = \frac{1}{8} [F_1(r) - F_2(r) \cos(4\theta)]. \quad (\text{A4})$$

Note that the angular dependence of this correlation function was obtained solely by checking how the atomic-level stress tensors are transformed under rotations (and mirror reflections). Thus, the physical properties of the liquid prescribe the r dependence of $F_{1,2}(r)$, but not the θ dependence in (A4).

APPENDIX B: ANOTHER DERIVATION OF THE EXPRESSION FOR $\langle \sigma_i^{xy} \sigma_j^{xy} \rangle$

Let us suppose that the first eigenvectors of atoms i and j form angles φ_i and φ_j with the \hat{x} axis of our reference coordinate frame. See Fig. 2.

From (10) the product $\sigma_i^{xy} \sigma_j^{xy}$ in our reference coordinate frame has the form:

$$\sigma_i^{xy} \sigma_j^{xy} = (1/4)(\lambda_i^1 - \lambda_i^2)(\lambda_j^1 - \lambda_j^2) \sin(2\varphi_i) \sin(2\varphi_j). \quad (\text{B1})$$

Note that the dependence on angles φ_i and φ_j appears in (B1) from the rotations (10) of the stresses from the coordinate frames of their eigenvectors into our reference coordinate frame. Thus, dependence of (B1) on φ_i and φ_j reflects transformational properties of the stress tensors under rotations.

We then, using Fig. 2, express angles φ_i and φ_j through the angles ψ_{ij} , ψ_{ji} , and θ_{ij} : $\varphi_i = \psi_{ij} + \theta_{ij}$ and $\varphi_j = \psi_{ji} + \theta_{ij}$. Substitution of these expressions for φ_i and φ_j into (B1) (with the following averaging) leads to:

$$\langle \sigma_i^{xy} \sigma_j^{xy} \rangle_{\theta_{ij}=\theta} = (1/8) [F_1 - F_2 \cos(4\theta) + F_3 \sin(4\theta)], \quad (\text{B2})$$

where F_1 , F_2 , and F_3 are given by expressions (23)–(25). Note that (40) is identical to (22) and (A4).

Since the dependence of (B1) on φ_i and φ_j appeared from the rotational properties of the stress tensors the dependence of (40) and (22) on θ also reflects the rotational properties of the stress tensors.

APPENDIX C: ESHELBY'S STRESS FIELD IN THE DIRECTIONAL FRAME FOR A CASE OF SHEAR DEFORMATION OF A CIRCULAR INCLUSION. FUNCTIONS $\mathcal{F}_1^e, \mathcal{F}_2^e, \mathcal{F}_3^e$

In this section we derive the expressions relating the eigenvalues and eigenvectors of the stress fields in the inclusion and in the matrix for a particular case of unconstrained traceless shear strain applied to the inclusion. Then we calculate functions $\mathcal{F}_1^e, \mathcal{F}_2^e, \mathcal{F}_3^e$ for the considered example. We start from the known formulas for Eshelby's stress field [18,29–32]. In particular, we use the expressions provided in Ref. [32].

We consider a particular case of unconstrained shear strain applied to the initially circular inclusion:

$$\epsilon_{\alpha\beta}^* = \epsilon^*(2\hat{n}_\alpha\hat{n}_\beta - \delta_{\alpha\beta}), \quad (\text{C1})$$

where \hat{n} is a 2D unit vector that determines the direction of deformation:

$$\hat{n}_x = \cos(\psi_{IJ}), \quad \hat{n}_y = \sin(\psi_{IJ}). \quad (\text{C2})$$

The expression for the final stress field in the inclusion (in the absence of external driving force) from formula (14) of Ref. [32] is

$$\sigma_{\alpha\beta}^I = g\epsilon_{\alpha\beta}^*, g \equiv \frac{-\mathcal{E}}{4(1-\nu^2)}, \quad (\text{C3})$$

where \mathcal{E} is the Young's modulus, while ν is the Poisson's ratio. The eigenvalues and eigenvectors of the stress tensor (C3) can be easily found:

$$\lambda_I^1 = +g\epsilon^*, \quad V_I^1 = [+\cos(\psi_{IJ}), +\sin(\psi_{IJ})], \quad (\text{C4})$$

$$\lambda_I^2 = -g\epsilon^*, \quad V_I^2 = [-\sin(\psi_{IJ}), +\cos(\psi_{IJ})]. \quad (\text{C5})$$

The expression for the final stress field in the matrix, according to formula (A25) of Ref. [32], is

$$\sigma_{\alpha\beta}^M = -g\epsilon^* \left\{ [\dots]_{\alpha\beta} - 4\nu \left(\frac{a^2}{r^2} \right) \left[\frac{2(\hat{n}\mathbf{r})^2}{r^2} - 1 \right] \delta_{\alpha\beta} \right\}, \quad (\text{C6})$$

where

$$\begin{aligned} [\dots]_{\alpha\beta} = & -4(1/\tilde{r})^2 \{ (1-2\nu) + (1/\tilde{r})^2 \} \\ & \cdot \{ (\hat{n}\hat{\mathbf{r}})(\hat{n}_\alpha\hat{r}_\beta + \hat{n}_\beta\hat{r}_\alpha) - \hat{r}_\alpha\hat{r}_\beta \} \\ & + (1/\tilde{r})^2 \{ 2(1-2\nu) + (1/\tilde{r})^2 \} \{ 2\hat{n}_\alpha\hat{n}_\beta - \delta_{\alpha\beta} \} \\ & - 4(1/\tilde{r})^2 \{ 1 - 2(1/\tilde{r})^2 \} \{ 2(\hat{n}\hat{\mathbf{r}})^2 - 1 \} \hat{r}_\alpha\hat{r}_\beta \\ & + 4(1/\tilde{r})^2 \{ 1 - (1/\tilde{r})^2 \} \\ & \cdot \{ (\hat{n}\hat{\mathbf{r}})(\hat{n}_\alpha\hat{r}_\beta + \hat{n}_\beta\hat{r}_\alpha) - 2(\hat{n}\hat{\mathbf{r}})^2\hat{r}_\alpha\hat{r}_\beta \} \\ & + 2(1/\tilde{r})^2 \{ 1 - (1/\tilde{r})^2 \} \{ 2(\hat{n}\hat{\mathbf{r}})^2 - 1 \} \delta_{\alpha\beta}. \end{aligned} \quad (\text{C7})$$

In (C7) $\tilde{r} \equiv r/a$. We are interested in the expression for the stress field in the coordinate frame associated with the direction from I to J . In this frame $\hat{\mathbf{r}} = (1,0)$, while $(\hat{n}\hat{\mathbf{r}}) = \cos(\psi_{IJ})$. Also note that $\hat{n}_x\hat{r}_x = \cos(\psi_{IJ})$ and $\hat{n}_y\hat{r}_x = \sin(\psi_{IJ})$, while $\hat{n}_x\hat{r}_y = 0$ and $\hat{n}_y\hat{r}_y = 0$. It is straightforward to obtain from (C7) the following expressions:

$$[\dots]_{xx} = \left(\frac{a}{r} \right)^2 \left\{ 2 - 3 \left(\frac{a}{r} \right)^2 \right\} \sin(2\psi_{IJ}), \quad (\text{C8})$$

$$[\dots]_{xx} = \left(\frac{a}{r} \right)^2 \left\{ -4(1-\nu) + 3 \left(\frac{a}{r} \right)^2 \right\} \cos(2\psi_{IJ}), \quad (\text{C9})$$

$$[\dots]_{yy} = \left(\frac{a}{r} \right)^2 \left\{ 4\nu - 3 \left(\frac{a}{r} \right)^2 \right\} \cos(2\psi_{IJ}). \quad (\text{C10})$$

Using expressions (C6) and (C8)–(C10) for the stress field in the matrix we get:

$$\sigma_{xy}^M(J) = -g\epsilon^* \left(\frac{a}{r} \right)^2 \left\{ 2 - 3 \left(\frac{a}{r} \right)^2 \right\} \sin(2\psi_{IJ}), \quad (\text{C11})$$

$$\sigma_{xx}^M(J) = -g\epsilon^* \left(\frac{a}{r} \right)^2 \left\{ -4 + 3 \left(\frac{a}{r} \right)^2 \right\} \cos(2\psi_{IJ}), \quad (\text{C12})$$

$$\sigma_{yy}^M(J) = -g\epsilon^* \left(\frac{a}{r} \right)^2 \left\{ -3 \left(\frac{a}{r} \right)^2 \right\} \cos(2\psi_{IJ}). \quad (\text{C13})$$

Formulas (C11)–(C13) give the components of the stress tensor at point J in the frame associated with the direction \mathbf{r}_{IJ} . These stress components are expressed in terms of the magnitude of the inclusion's unconstrained strain, i.e., ϵ^* , and the direction of the strain, i.e., ψ_{IJ} , with respect to the direction \mathbf{r}_{IJ} .

The eigenvalues, λ_M^1 and λ_M^2 , and eigenvectors, \mathbf{V}_M^1 and \mathbf{V}_M^2 , of the stress matrix in the frame associated with \mathbf{r}_{IJ} can now be found:

$$\lambda_M^1 = -g\epsilon^* \left(\frac{a}{r} \right)^2 \left\{ -4[\cos(\psi_{IJ})]^2 + 3 \left(\frac{a}{r} \right)^2 \right\}, \quad (\text{C14})$$

$$\lambda_M^2 = -g\epsilon^* \left(\frac{a}{r} \right)^2 \left\{ +4[\sin(\psi_{IJ})]^2 - 3 \left(\frac{a}{r} \right)^2 \right\}, \quad (\text{C15})$$

$$\mathbf{V}_M^1 = [+\cos(\psi_{IJ}), -\sin(\psi_{IJ})], \quad (\text{C16})$$

$$\mathbf{V}_M^2 = [+\sin(\psi_{IJ}), +\cos(\psi_{IJ})]. \quad (\text{C17})$$

It follows from (C4), (C5) and (C16), (C17) that:

$$\psi_{JI} = -\psi_{IJ}. \quad (\text{C18})$$

Now we are in a position to write expressions for the functions $\mathcal{F}_1^e, \mathcal{F}_2^e, \mathcal{F}_3^e$ from (41)–(43). It follows from (C4), (C5) that for the inclusion we have:

$$\lambda_I^1 - \lambda_I^2 = 2g\epsilon^*, \quad (\text{C19})$$

while for the matrix from (C14), (C15):

$$\lambda_M^1 - \lambda_M^2 = -2g\epsilon^* \left(\frac{a}{r} \right)^2 \left\{ -2 + 3 \left(\frac{a}{r} \right)^2 \right\}. \quad (\text{C20})$$

Thus,

$$f_o(r) \equiv (\lambda_I^1 - \lambda_I^2)(\lambda_M^1 - \lambda_M^2) \quad (\text{C21})$$

$$= -(2g\epsilon^*)^2 \left(\frac{a}{r} \right)^2 \left\{ -2 + 3 \left(\frac{a}{r} \right)^2 \right\}. \quad (\text{C22})$$

By taking into account that $\psi_{JI} = -\psi_{IJ}$ from (41)–(43) we get:

$$\mathcal{F}_1^e = f_o(r) \cos(4\psi_{IJ}), \quad \mathcal{F}_2^e = f_o(r), \quad \mathcal{F}_3^e = 0. \quad (\text{C23})$$

In order to find the average values of the functions above it is necessary to average them over all values of ψ_{IJ} . Note that the function \mathcal{F}_1^e averages to zero. This fact is

of interest since in liquids (\mathcal{F}_1^e) is not zero and it is the function that is the most directly related to viscosity, see Fig. 5.

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the relevant stress components of (I) and (J) in the reference Cartesian coordinate frame is taken.

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6(a) of Ref. [14] can be reproduced using function $F_5(r)$ and formula (26) of this paper. Function $F_4(r)$ that enters into (26) is zero. Figure 5(c) of Ref. [14] can be reproduced using function $F_1(r)$ and formula (29) of this paper. Function $F_6(r)$ that enters into (29) is zero. Figure 6(b) shows function $\langle p_i p_j \rangle$ which is essentially equivalent to the function G_{pp} from (31). Figure 6(c) of Ref. [14] can be reproduced using function $F_5(r)$ and formula $\langle p_i(\sigma_j^{xx} - \sigma_j^{yy}) \rangle = F_5(r) \cos(2\theta_{ij})$ that can be easily derived.