Approximate analytical description of the nonaffine response of amorphous solids

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An approximation scheme for model disordered solids is proposed that leads to the fully analytical evaluation of the elastic constants under explicit account of the inhomogeneity (nonaffinity) of the atomic displacements. The theory is in quantitative agreement with simulations for central-force systems and predicts the vanishing of the shear modulus at the isostatic point with the linear law $\mu \sim (z - 2d)$, where z is the coordination number. The vanishing of rigidity at the isostatic point is shown to be a consequence of the canceling out of positive affine and negative nonaffine terms.

I. INTRODUCTION

Disordered or amorphous solids represent a great part of ordinary matter (e.g., glass), including biological matter (e.g., cytoskeletal networks).[1–3] Yet the relationship between rigidity and disorder has remained elusive and no theory has hitherto proved able to correctly describe their elastic constants. The rigidity of disordered solids is also intimately related to the fundamentally unsolved problem of the glass transition.[4] Elastic rigidity in supercooled liquids emerges from the fluid state at the glass transition without any detectable lowering of the symmetry (apart from translational and replica symmetry-breaking), as opposed to what happens in "ordinary" liquid-solid phase transitions.[5] In recent years, it has been recognized that the intrinsic "softness" of disordered solids is related to the *nonaffinity* of the atomic displacements:[1,4,6,7] the atoms in a strained disordered solid are *not* displaced *proportionally* to the global strain. The calculation of the contribution to rigidity due to such nonaffine displacements poses formidable difficulties because it requires the analytical knowledge of the eigenmodes of the dynamical or Hessian matrix of the system,[8,9] which is a sparse random matrix. The simplest disordered solids where nonaffinity is supposed to play a significant role are those with nearest-neighbor central-force interactions.[9,10] In these systems, it is well-known that the shear modulus μ vanishes at the isostatic point where each particle has an average number of nearest neighbors (mechanical contacts) $z = 2d$.[9,10] Although it seems reasonable from constraint-counting arguments that rigidity is lost when $z = 2d$,[11] the linear law $\mu \sim (z - 2d)$ observed in simulation studies of completely disordered solids[12] has remained unexplained and the physics behind it represents a long-standing problem[9,10] where the role of nonaffinity is unclear. Further, the well-documented inadequacy of affine theories to describe the elasticity and transport properties of amorphous materials calls for an improved theory beyond the affine approximation.[13] In this paper, we propose an approximation scheme that gives a well-defined deterministic limit for the nonaffine contributions to the elastic constants. This leads to a fully analytical description of the elastic constants that accounts for the microscopic nonaffinity of the atomic displacements.

II. FORMALISM

In the following, Roman indices are used to label atoms while Greek indices are used to label Cartesian components. The summation convention over repeated indices holds throughout for Greek indices. Bold characters denote vectors in dN-dimensional space (N is the total number of atoms). We closely follow the notation of Lemaitre and Maloney[8] and we start our analysis from the definition of a Bravais cell for the disordered lattice. The cell is described by three Bravais vectors and thus by a matrix $\underline{h}$. The potential depends on the particle position $\underline{r}_i$ and on the shape of the cell, which enforces boundary conditions. Macroscopically imposed deformations of the cell are described by changes in the Bravais vectors through a linear map $\underline{F} = \underline{h}\underline{\mathring{h}}^{-1}$ and the relation $\underline{h} = \underline{F}\,\underline{\mathring{h}}$. We denote quantities in the reference frame, as well as quantities that are measured with respect to the reference frame, with a circle. Accordingly, the unit cell in the reference configuration is given through the matrix $\underline{\mathring{h}}$. In the language of continua, $\underline{F}$ is the deformation gradient tensor. The position of atom i after an affine deformation is given by

$$r_i^\alpha = F_{\alpha\beta} R_i^\beta. \tag{1}$$

As a unique exception to the ring notation, we denote with $\underline{R}_i$ the position of the atoms in the reference cell. This relation illustrates the definition of affine displacement as atom i is displaced *proportionally* to the external deformation. The position of the atom after a strain is denoted by $r_i^\alpha(\underline{F})$ and differs from Eq. (1) if the nonaffine displacement is not zero. It is useful to introduce the particle position $\underline{\mathring{r}}_i$ for an atom that undergoes both affine and nonaffine displacements, defined by

$$r_i^\alpha(\underline{F}) = F_{\alpha\beta}\,\mathring{r}_i^\beta(\underline{F}). \tag{2}$$

Remark also that $\underline{R}_i = \underline{\mathring{r}}_i(0)$. Hence, for an affine displacement the position vector $\underline{\mathring{r}}_i$ is kept fixed while the new position is determined by $\underline{F}$. Any additional (*nonaffine*) displacement is thus parametrized for a given strain in terms of $\underline{\mathring{r}}_i(\underline{F})$. With these definitions we can express the potential $\mathcal{U}(\{\underline{r}_i\},\underline{F})$ in the coordinates of the reference frame as $\mathring{\mathcal{U}}(\{\underline{\mathring{r}}_i\},\underline{F})$, which is defined by

$$\mathring{\mathcal{U}}(\{\underline{\mathring{r}}_i\},\underline{F}) = \mathcal{U}(\{\underline{F}\,\underline{\mathring{r}}_i\},\underline{F}). \tag{3}$$

It is convenient to introduce the Cauchy-Green strain tensor $\underline{\underline{\eta}} = \frac{1}{2}(\underline{\underline{F}}^\top \underline{\underline{F}} - \underline{\underline{I}})$ to describe the deformations since the elastic constants are defined in terms of second derivatives in η. The deformation is completely described by this tensor since the total internal energy of the solid being deformed can be expressed as $\mathcal{U}(\{|\underline{r}_{ij}|\})$, i.e., as a functional of the set $\{r_{ij}\}$ of relative distances between atoms $r_{ij} = |\underline{r}_{ij}| = |\underline{r}_i - \underline{r}_j|$, and $\underline{\underline{\eta}}$ describes affine transformations in terms of relative interatomic distances according to $|\underline{\underline{F}} \, \underline{R}_{ij}|^2 = |\underline{R}_{ij}|^2 + \underline{R}_{ij}^\top \underline{\underline{\eta}} \, \underline{R}_{ij}$.

A homogeneous strain of the cell $\underline{\underline{F}}$ will first bring each atom to its affine position $r_i^\alpha = F_{\alpha\beta} R_i^\beta$. In this affine position, the total force acting on atom i is in general not zero since the neighboring atoms may exert a nonvanishing force field due to their affine motion. This is especially true for disordered solids where the nearest neighbors are placed at random around atom i so that they transmit unbalanced forces to i (in an ordered lattice, the transmitted forces balance by symmetry such that this effect is often negligible). It is in response to these virtual forces that the atom undergoes an additional motion after it has been displaced affinely such that the energy released in the process reestablishes (local) mechanical equilibrium. Hence the system under reversible strain evolves adiabatically along a trajectory $\underline{r}(\eta)$ that minimizes the mechanical energy for a given strain $\underline{\underline{\eta}}$. If we denote by $\frac{\mathcal{D}}{\mathcal{D}\eta_{\kappa\chi}}$ the derivative with respect to adiabatic changes of the strain under the constraint of mechanical equilibrium, one obtains an equation of motion of the nonaffine displacement by differentiating the force $f_i^\alpha = -\frac{\partial \mathcal{U}}{\partial r_i^\alpha}$ evaluated in the true position (where it vanishes). In the limit $\underline{\underline{\eta}} \to 0$, the equation reads

$$\sum_j H_{ij}^{\alpha\beta} \left. \frac{\mathcal{D}\mathring{r}_j^\beta}{\mathcal{D}\eta_{\kappa\chi}} \right|_{\underline{\underline{\eta}}=0} = \Xi_{i,\kappa\chi}^\alpha, \tag{4}$$

where we have introduced the Hessian $H_{ij}^{\alpha\beta}$ and the affine force field $\Xi_{i,\kappa\chi}^\alpha$ given, respectively, by

$$H_{ij}^{\alpha\beta} = \left. \frac{\partial^2 \mathring{\mathcal{U}}}{\partial \mathring{r}_i^\alpha \partial \mathring{r}_j^\beta} \right|_{\underline{\underline{\eta}}=0} = \left. \frac{\partial^2 \mathcal{U}}{\partial r_i^\alpha \partial r_j^\beta} \right|_{\underline{\underline{\eta}}=0},$$

$$\Xi_{i,\kappa\chi}^\alpha = -\left. \frac{\partial^2 \mathring{\mathcal{U}}}{\partial \mathring{r}_i^\alpha \partial \eta_{\kappa\chi}} \right|_{\underline{\underline{\eta}}=0} = -\sum_j \left. \frac{\partial^2 \mathring{\mathcal{U}}}{\partial \mathring{r}_i^\alpha \partial \mathring{r}_j^\beta} \right|_{\underline{\underline{\eta}}=0} \frac{\partial F_{\beta\beta'}}{\partial \eta_{\kappa\chi}} R_j^{\beta'}. \tag{5}$$

The elastic constants are defined by

$$C_{\iota\xi\kappa\chi} = \frac{1}{\mathring{V}} \left. \frac{\partial^2 \mathcal{U}}{\partial \eta_{\iota\xi} \partial \eta_{\kappa\chi}} \right|_{\underline{\underline{\eta}}=0}. \tag{6}$$

To account for the nonaffine relaxation in the calculation of the elastic constants, the derivatives in Eq. (6) have to be taken along a trajectory of (locally) minimum energy. Following

Lemaitre and Maloney,[8] we obtain

$$C_{\iota\xi\kappa\chi} = \frac{1}{\mathring{V}} \left[\frac{\mathcal{D}}{\mathcal{D}\eta_{\iota\xi}} \left(\frac{\partial \mathring{\mathcal{U}}}{\partial \eta_{\kappa\chi}} + \frac{\partial \mathring{\mathcal{U}}}{\partial \mathring{r}_i} \frac{\mathcal{D}\mathring{r}_i}{\mathcal{D}\eta_{\kappa\chi}} \right) \right]_{\eta=0}$$

$$= \frac{1}{\mathring{V}} \left(\left. \frac{\partial^2 \mathcal{U}}{\partial \eta_{\iota\xi} \partial \eta_{\kappa\chi}} \right|_{\eta=0} + \left. \frac{\partial^2 \mathcal{U}}{\partial \mathring{r}_i \partial \eta_{\iota\xi}} \right|_{\eta=0} \left. \frac{\mathcal{D}\mathring{r}_i}{\mathcal{D}\eta_{\kappa\chi}} \right|_{\eta=0} \right)$$

$$= \frac{1}{\mathring{V}} \left. \frac{\partial^2 \mathcal{U}}{\partial \eta_{\iota\xi} \partial \eta_{\kappa\chi}} \right|_{\eta=0} - \frac{1}{\mathring{V}} \Xi_{i,\iota\xi} \left. \frac{\mathcal{D}\mathring{r}_i}{\mathcal{D}\eta_{\kappa\chi}} \right|_{\eta=0}$$

$$= C_{\iota\xi\kappa\chi}^A - C_{\iota\xi\kappa\chi}^{NA}, \tag{7}$$

where it is evident that the true elastic constant is given by the affine (Born-Huang) elastic constant $C_{\iota\xi\kappa\chi}^A$ corrected by the nonaffine term $-C_{\iota\xi\kappa\chi}^{NA}$. Following Lemaitre and Maloney,[8] and using Eq. (6) in Eq. (7), one derives the following expression for the nonaffine correction:

$$C_{\iota\xi\kappa\chi}^{NA} = \Xi_{i,\iota\xi}^\alpha (H_{ij}^{\alpha\beta})^{-1} \Xi_{j,\kappa\chi}^\beta > 0. \tag{8}$$

The last inequality in Eq. (8) is justified in view of the Hessian matrix being semipositive definite at mechanical equilibrium. Hence it follows that the correction due to the nonaffine relaxation, $-C_{\iota\xi\kappa\chi}^{NA} < 0$, necessarily gives a *negative* contribution to the total rigidity.[8]

III. APPROXIMATION SCHEME

A. The Cauchy bonded-network model

Let us consider the disordered Cauchy solid, defined by the following properties:[1] (i) atoms interact pairwise and only with their nearest neighbors; (ii) the interaction potential is a central-force harmonic potential; (iii) the reference state is *unstressed*, i.e., all springs (interatomic bonds) are relaxed in the minimum of the harmonic well; (iv) disorder is spatially decorrelated. The equivalence with a random network of harmonic springs is evident.[9,14] Hence, the total free energy is given by $\mathcal{U}(\{r_{ij}\}) = \sum_{\langle ij \rangle} V_{ij}(r_{ij})$, where the sum runs over all pairs of nearest neighbors $\langle ij \rangle$. The pair interaction potential is given by the harmonic potential $V(r_{ij}) = \frac{\kappa}{2}(r_{ij} - R_0)^2$. κ is the atomic force constant and R_0 is the interatomic distance at rest in the reference frame. Under these conditions, the Hessian matrix becomes

$$H_{ij}^{\alpha\beta} = \delta_{ij} \sum_s \kappa c_{is} n_{is}^\alpha n_{is}^\beta - (1 - \delta_{ij}) \kappa c_{ij} n_{ij}^\alpha n_{ij}^\beta, \tag{9}$$

where we used the identity $\partial/\partial \underline{r}_{ij} = \underline{n}_{ij} \partial/\partial r_{ij}$, with $\underline{n}_{ij} = \underline{r}_{ij}/r_{ij}$. Further, c_{ij} is the (random) occupancy matrix with $c_{ij} = 1$ if i and j are nearest neighbors and $c_{ij} = 0$ otherwise. c_{ij} is a matrix where each row and each column have on average z elements equal to 1 distributed randomly under the constraint that the matrix be symmetric. Using this form of the Hessian, one obtains the affine part of the elastic constant as

$$C_{\iota\xi\kappa\chi}^A = \frac{R_0^2 \kappa}{2\mathring{V}} \sum_{ij} c_{ij} n_{ij}^\iota n_{ij}^\xi n_{ij}^\kappa n_{ij}^\chi, \tag{10}$$

which is the well-known Born-Huang formula.[1,7,8] Further, we also obtain a microscopic expression for the affine force field from Eq. (5) as $\Xi_{i,\kappa\chi}^\alpha = -\sum_j R_{ij} \kappa c_{ij} n_{ij}^\alpha n_{ij}^\kappa n_{ij}^\chi$. We can now turn to the nonaffine part of the elastic stiffness, $C_{\iota\xi\kappa\chi}^{NA}$.

The Hessian is a $dN \times dN$ symmetric semipositive-definite matrix with d eigenvalues equal to zero that are due to the global translational invariance of the solid. Equation (4) can be solved by normal mode decomposition, which leads to[8]

$$C_{\iota\xi\kappa\chi}^{NA} = \frac{1}{\overset{\circ}{V}} \sum_{\substack{k \\ \lambda_k \neq 0}} \frac{(\underline{\Xi}_{\iota\xi}, \underline{v}_k)(\underline{\Xi}_{\kappa\chi}, \underline{v}_k)}{\lambda_k}, \qquad (11)$$

where $\underline{v}_k$ are the eigenvectors of the Hessian (which are orthogonal since the Hessian is symmetric), λ_k denotes the corresponding eigenvalues, and $(,)$ denotes the normal scalar product on $\mathbb{R}^{dN}$. In the next section, we shall evaluate the deterministic limit of Eq. (11), which is a self-averaging quantity.[8]

B. The approximate Hessian and the affine field projection on its eigenmodes

A rigorous derivation requires one to first determine the eigenmodes $\underline{v}_k$ of the Hessian matrix given by Eq. (9) to calculate the projection on them of the affine fields in Eq. (11). Thereafter, the average over the disorder is taken to get the thermodynamic limit of $C_{\iota\xi\kappa\chi}^{NA}$. However, the Hessian is a random matrix and both $\underline{v}_k$ and λ_k depend on the realization of disorder. Also, being the Hessian sparse, there are no analytical forms even for its statistical spectral distributions. Nevertheless, the deterministic limit of Eq. (11) can be calculated analytically within the following approximation that we propose here.

Our approximation consists in performing a disorder average of the orientation-dependent part of the Hessian first and then using the result to calculate the eigenmodes, their inner products with the affine fields, and finally the nonaffine correction. Inverting the sequence of "calculating" and "averaging" is sometimes referred to as an effective-medium approximation and is not at all unusual in dealing with disordered systems[15] since it is often the only strategy to keep the treatment analytical. Using some averaged form of the Hessian matrix necessarily implies sacrificing some details of the vibrational spectrum. This problem is addressed in Secs. IV B and IV C, where we show what details are lost and we study the validity and limitations of the approximation.

In $d = 3$ it is $\underline{n}_{ij} = (\cos\phi_{ij}\sin\theta_{ij}, \sin\phi_{ij}\sin\theta_{ij}, \cos\theta_{ij})$ and the pair of angles ϕ_{ij} and θ_{ij} univocally specifies the orientation of the bond $\langle ij \rangle$. The orientation-dependent factors in the Hessian, $n_{ij}^{\alpha} n_{ij}^{\beta}$ in Eq. (9), for a large system with uncorrelated isotropic disorder (where every bond can take any orientation in the solid angle with the same probability $1/4\pi$), can be replaced with its isotropic (angular) average, i.e., $n_{ij}^{\alpha} n_{ij}^{\beta} \Rightarrow \delta_{\alpha\beta}/d$. Within this approximation, the Hessian becomes

$$H_{ij}^{\alpha\beta} = \frac{\kappa}{d} \left(\delta_{ij} \sum_j c_{ij} - (1 - \delta_{ij})c_{ij} \right) \delta_{\alpha\beta}. \qquad (12)$$

Note that this is still a sparse random matrix because of the positional disorder in the random coefficients c_{ij}. According to Eq. (12), let us define $\underline{\underline{H}} = \underline{\underline{\tilde{H}}} \otimes \underline{\underline{I}}$, where $\underline{\underline{I}}$ is the $d \times d$ identity matrix (which represents $\delta_{\alpha\beta}$) and $\underline{\underline{\tilde{H}}}$ is the matrix that multiplies $\delta_{\alpha\beta}$ in Eq. (12). Denoting with $\{\underline{a}_q\}_{q=1,...,N}$

the set of eigenvectors of $\underline{\underline{\tilde{H}}}$, which is an orthonormal basis (ONB) of $\mathbb{R}^N$, and with $\{\underline{e}_l\}_{l=1,...,d}$ the standard Cartesian basis of $\mathbb{R}^d$, it follows that $(\underline{\underline{\tilde{H}}} \otimes \underline{\underline{I}})(\underline{a} \otimes \underline{e}) = \lambda(\underline{a} \otimes \underline{e})$ and thus the dN-dimensional set $\overline{\{\underline{a}_q \, \underline{e}_l\}}_{q=1,...,N, l=1,...,d}$ is an ONB of $\underline{\underline{H}}$ as given by Eq. (12). This allows us to write (with $\underline{v} = \underline{a} \, \underline{e}_l$ for some $\underline{a} \in \{\underline{a}_q\}_{q=1,...,N}$)

$$(\underline{\Xi}_{\iota\xi}, \underline{v})(\underline{\Xi}_{\kappa\chi}, \underline{v}) = \left(\sum_r^N a_r \underline{\Xi}_{r,\iota\xi} \, \underline{e}_l \right) \left(\sum_r^N a_r \underline{\Xi}_{r,\kappa\chi} \, \underline{e}_l \right)$$

$$= \kappa^2 R_0^2 \sum_{r \, s \, r' \, s'} \left\{ (a_r a_{r'} \, c_{rs} c_{r's'}) \qquad (13) \right.$$

$$\left. \times \left(n_{rs}^l n_{rs}^\iota n_{rs}^\xi n_{r's'}^l n_{r's'}^\kappa n_{r's'}^\chi \right) \right\}.$$

With our isotropic approximation, we replace the orientation-dependent terms with their isotropic angular-averaged values, which gives $n_{rs}^l n_{rs}^\iota n_{rs}^\xi n_{r's'}^l n_{r's'}^\kappa n_{r's'}^\chi = (\delta_{rr'}\delta_{ss'} - \delta_{rs'}\delta_{sr'})B_{l,\iota\xi\kappa\chi}$, where the $B_{l,\iota\xi\kappa\chi}$ are geometric coefficients resulting from the angular average. For $d = 3$ and 2 they are as follows:

l	$d = 3$				$d = 2$		
	x	y	z	$\sum_l$	x	y	$\sum_l$
$B_{l,xxxx}$	$\frac{1}{7}$	$\frac{1}{35}$	$\frac{1}{35}$	$\frac{1}{5}$	$\frac{5}{16}$	$\frac{1}{16}$	$\frac{3}{8}$
$B_{l,xyxy}$	$\frac{1}{35}$	$\frac{1}{35}$	$\frac{1}{105}$	$\frac{1}{15}$	$\frac{1}{16}$	$\frac{1}{16}$	$\frac{1}{8}$
$B_{l,xxyy}$	$\frac{1}{35}$	$\frac{1}{35}$	$\frac{1}{105}$	$\frac{1}{15}$	$\frac{1}{16}$	$\frac{1}{16}$	$\frac{1}{8}$

$$(14)$$

Substituting in Eq. (11), we obtain

$$(\underline{\Xi}_{\iota\xi}, \underline{v})(\underline{\Xi}_{\kappa\chi}, \underline{v}) = \kappa^2 R_0^2 \, B_{l,\iota\xi\kappa\chi}$$

$$\times \left(\sum_{r \, s} a_r^2 \, c_{rs} c_{rs} - \sum_{r \, s} a_r a_s \, c_{rs} c_{sr} \right)$$

$$= \kappa^2 R_0^2 \, B_{l,\iota\xi\kappa\chi} \frac{d}{\kappa} \sum_{rs}^N a_r a_s \tilde{H}_{rs}, \qquad (15)$$

where we used that $c_{rs}^2 = c_{rs}c_{sr} = c_{rs}$ and the identities $\sum_r^N a_r^2 \sum_s c_{rs} - \sum_{rs} a_r a_s c_{rs} = \sum_{rs}^N a_r a_s [(\sum_j^N c_{rj})\delta_{rs} - c_{rs}(1 - \delta_{rs})] = \frac{d}{\kappa} \sum_{rs}^N a_r a_s \tilde{H}_{rs}$. Recalling that $\sum_s^N \tilde{H}_{rs} a_s = \lambda a_r$, we obtain $(\underline{\Xi}_{\iota\xi}, \underline{v}_k)(\underline{\Xi}_{\kappa\chi}, \underline{v}_k) = d\kappa R_0^2 \lambda_k B_{l,\iota\xi\kappa\chi}$.

IV. RESULTS AND DISCUSSION

A. Elastic moduli

We have shown that within the isotropic approximation of the Hessian, Eq. (12), the nonaffine part of the elastic stiffness, Eq. (11), has the following thermodynamic limit:

$$\langle C_{\iota\xi\kappa\chi}^{NA} \rangle = \frac{1}{\overset{\circ}{V}} \sum_{q=1}^N \sum_{l=1}^d \frac{d\kappa R_0^2 \lambda_q \, B_{l,\iota\xi\kappa\chi}}{\lambda_q}$$

$$= d\frac{N}{\overset{\circ}{V}} \kappa R_0^2 \sum_{l=1}^d B_{l,\iota\xi\kappa\chi}. \qquad (16)$$

The affine part of the elastic constants for the disordered Cauchy solid can be obtained by performing the disorder average of Eq. (10), $\langle C_{\iota\xi\kappa\chi}^A \rangle$, where $\langle \, \rangle$ denotes the angular average, and we always use the isotropic distribution of the
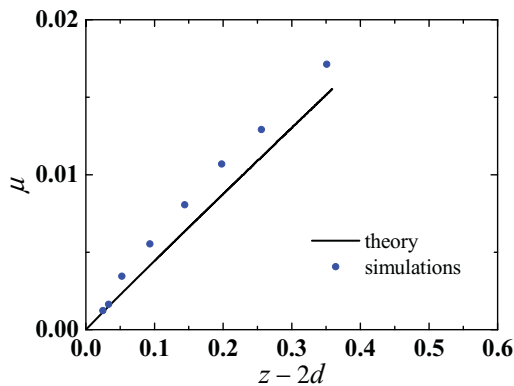
FIG. 1. (Color online) The theoretical predictions for the shear modulus, Eq. (17), and the simulation results of Ref. 12. No fitting parameter is used in the comparison. $\kappa = 1$, $R_0 = 1$.

bond orientations. In $d = 3$, we thus obtain $\mu^A = \langle C^A_{xyxy}\rangle = \frac{1}{30}\frac{N}{V}\kappa z R_0^2$ for the shear modulus and $K^A = \frac{1}{3}(\langle C^A_{xxxx}\rangle + 2\langle C^A_{xxyy}\rangle) = \frac{1}{18}\frac{N}{V}\kappa z R_0^2$ for the bulk modulus. Therefore, using these affine moduli together with the coefficients of Eq. (14) and with Eq. (16), we derive expressions for the shear and bulk modulus of the $d = 3$ disordered Cauchy solid, respectively, as

$$\mu = \mu^A - \mu^{NA} = \frac{1}{30}\frac{N}{V}\kappa R_0^2(z - 6),$$
$$K = K^A - K^{NA} = \frac{1}{18}\frac{N}{V}\kappa R_0^2(z - 6). \quad (17)$$

For $d = 2$, we obtain

$$\mu = \mu^A - \mu^{NA} = \frac{1}{16}\frac{N}{V}\kappa R_0^2(z - 4),$$
$$K = K^A - K^{NA} = \frac{5}{48}\frac{N}{V}\kappa R_0^2(z - 4). \quad (18)$$

Generalizing this result to arbitrary space dimensions gives the following scaling for the moduli in d dimensions:

$$\mu \sim K \sim (z - 2d). \quad (19)$$

The predictions of Eq. (17) for the shear modulus, without fitting parameters, can be compared with the simulations of Ref. 12 of $d = 3$ disordered packings of (monodisperse) compressible spheres interacting via harmonic repulsion in Fig. 1. In the simulations, the frictionless spheres at $T = 0$ are slightly compressed to packing fractions ϕ just above the so called jamming point at $\phi_J = 0.64$, which is also an isostatic point with $z_J = 2d$.[9,12] Further, the jamming point is a zero-stress point,[12] and the effect of stress on the global rigidity is therefore small. However, it seems from this comparison, and from our results, that stresses, in general, are not likely to affect the *qualitative* behavior of the *global* rigidity as they play no role in arriving at the fundamental scaling law Eq. (19).

Finally, we note that the vanishing of K at the isostatic point predicted by our theory does not agree with the scaling of K observed in soft-sphere packings where it remains finite at $z_J = 2d$.[12,14] It agrees, however, with the behavior of random networks where K vanishes linearly at $z_J = 2d$.[14] The reason for this might be tentatively identified with the fact that in

our theory, just like in networks, excluded volume effects are irrelevant, whereas they are important in packings.[14]

B. Vibrational density of states and validity of the approximation

The isotropic Hessian matrix introduced for the calculation of the nonaffine contribution to the elastic moduli can be used to obtain the density of vibrational states (DOS) numerically. Recall that the approximate Hessian is given by the following random matrix:

$$H^{\alpha\beta}_{ij} = \frac{\kappa}{3}\left(\delta_{ij}\sum_j c_{ij} - (1 - \delta_{ij})c_{ij}\right)\delta_{\alpha\beta}.$$

The Hessian is defined by the coefficients c_{ij}, which depend on the realization σ and are, therefore, random variables. The eigenvalues and thus the eigenvalue distribution of a random matrix are also random quantities. The explicit calculation of the eigenvalues as functions of the matrix elements is not possible. The approach to the eigenvalue problem in random-matrix theory makes use of the self-averaging assumption that the eigenvalue distribution becomes deterministic in the limit of an infinite system size.[16] As analytical solutions for the eigenvalue distribution in our case are not possible (due to the sparseness of the Hessian), we resort to a numerical analysis assuming that the self-averaging property holds. Therefore, we can define a limiting eigenvalue distribution $\rho(\lambda)$ as follows:

$$\lim_{N\to\infty}\langle\rho^N(\lambda)\rangle_\sigma = \rho(\lambda).$$

Then we have for all realizations σ that

$$\lim_{N\to\infty}\rho_N(\lambda)[\sigma] = \rho(\lambda).$$

Remark that for a finite N, the eigenvalues distribution is discrete and given by

$$\rho^N(\lambda)[\sigma] = \frac{1}{N}\sum_{i=1}^{N}\delta(\lambda - \lambda_i),$$

where δ is the Dirac delta function. In the limit $N\to\infty$, the set of eigenvalues has infinite elements, and the distribution becomes continuous. The factor $1/N$ is necessary to normalize the density given that the normalization condition is $\int_0^\infty\rho(\lambda)\,d\lambda = 1$. To analyze numerically the eigenvalue distribution, we calculate the eigenvalues sets $\{\lambda_i\}_r$ for large systems ($N \simeq 10\,000$). Then we create an histogram of the eigenvalues set and we fit it with a continuous curve that approximates the limiting eigenvalue distribution. The eigenvalue distribution is usually described in terms of the vibrational DOS, which we denote as $D(\omega)$, where ω is the vibrational frequency. The latter is related to the eigenvalue distribution by the change of variables

$$\lambda \to \omega = \sqrt{\frac{\lambda}{m}} \quad \text{and its inverse} \quad \omega \to \lambda = m\omega^2.$$

Hence, with $d\omega = \frac{1}{2\sqrt{m\lambda}}\,d\lambda$ and $d\lambda = 2m\omega\,d\omega$ we get that the density distributions of λ and ω are related by

$$D(\omega) = \rho(m\omega^2)\,2\omega \quad \text{and} \quad \rho(\lambda) = \frac{D\left(\sqrt{\frac{\lambda}{m}}\right)}{2\sqrt{m\lambda}}.$$
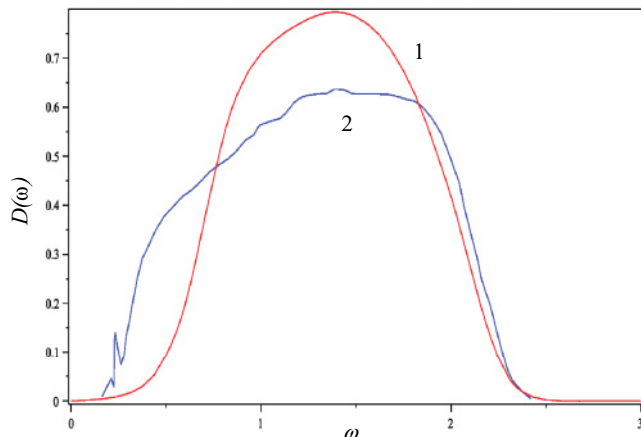
FIG. 2. (Color online) 1 (red), The DOS calculated for a system of $N = 10\,000$ particles according to Eq. (12) (solid line) and $z = 8$. 2 (blue), Simulations for repulsive unstressed harmonic packings of Ref. 17 with $\phi - \phi_c = 0.1$, which corresponds to $z - 6 \simeq 2$.

We compare the so obtained DOS from Eq. (12) with the DOS from simulations of unstressed harmonic packings[17] where $\phi - \phi_c = 0.1$ ($\phi_c \approx 0.64$ is the jamming packing fraction), corresponding to $z - 6 \simeq 2$. The comparison is shown in Fig. 2. While the upper end and the main features of the spectrum (width and average) are quite well reproduced, this was somewhat expected because sacrificing information about the bond orientations does not change the single-particle parameters (of order $\sqrt{\kappa/\omega}$) that dominate the highly (Anderson) localized high-ω modes. On the other hand, it is now clear from this comparison what details of the vibrational spectrum were lost in our isotropic approximation of the Hessian, Eq. (12): the spectrum calculated with the isotropic Hessian is significantly depleted of low-frequency modes whose density is significantly underestimated in comparison with the DOS of the packings. This is probably related to the anisotropic character of the local correlations between particles in the packings (ultimately related to excluded-volume and static effects), which play an important role in the low-frequency modes and which are lost in the approximation. This observation hints again at the possibility that overall our model describes random networks (where excluded volume is absent) better than sphere packings. The relationship between these observations and the excluded-volume effects should be more systematically investigated in future studies. This was already noted in relation to the bulk modulus prediction of our theory, which agrees indeed with the random-network scaling ($K \sim z - 6$), though not with the sphere packing one, for arguably similar reasons.

In view of these considerations, it is natural to ask why our theory, which seems to underestimate the low-ω modes, still yields a correct prediction of the shear modulus for sphere packings. This question is related to the issue of the role played by the low-frequency modes in the nonaffine response. While this issue is a very open and unsolved one in our current understanding of amorphous solids,[10] a tentative, and certainly incomplete, answer to this deep question is proposed in the next section.

C. On the role of low-frequency modes in the nonaffine response

To assess the relative importance of different regimes of the vibrational spectrum in the nonaffine elastic response, it is instructive to rewrite the elastic moduli with the nonaffine correction in the continuous frequency domain. According to the nonaffine linear-response formalism of Lemaitre and Maloney,[8] in the thermodynamic limit one has

$$\langle C_{\iota\xi\kappa\chi} \rangle = \langle C^A_{\iota\xi\kappa\chi} \rangle - \int_0^\infty \frac{D(\omega)\Gamma_{\iota\xi\kappa\chi}(\omega)}{m\omega^2} d\omega, \qquad (20)$$

where the correlators on the frequency shells are defined by

$$\Gamma_{\iota\xi\kappa\chi}(\omega) = \langle (\underline{\Xi}_{\iota\xi}, \mathbf{v}_k)(\underline{\Xi}_{\kappa\chi}, \mathbf{v}_k) \rangle_{\omega_k \in [\omega, \omega + d\omega]}. \qquad (21)$$

The function $\Gamma_{\iota\xi\kappa\chi}(\omega)$ thus represents the projection of the affine fields on the frequency shells, and its magnitude gives the importance of the contribution of each frequency shell to the nonaffine response. From Eq. (20) it is evident first of all that in the zero-frequency limit $\omega \to 0$ the moduli diverge to minus infinity, i.e., $\langle C_{\iota\xi\kappa\chi} \rangle \to -\infty$, unless either $D(\omega = 0) = 0$ or $\Gamma_{\iota\xi\kappa\chi}(\omega) = 0$. At the isostatic or jamming point of sphere packings, one has that the DOS develops soft modes with $D(\omega = 0) \neq 0$. As the nonaffine linear formalism is an exact theory, it is then strictly necessary that

$$\lim_{\omega \to 0} \Gamma_{\iota\xi\kappa\chi}(\omega) = 0, \qquad (22)$$

i.e., the zero-frequency modes must not contribute to the nonaffine response. This is what has been observed indeed in the numerical simulations of Ref. 8 where, in the case of a Lennard-Jones glass, the function $\Gamma_{\iota\xi\kappa\chi}(\omega)$ measured in the simulations goes to zero at $\omega = 0$. Furthermore, in the same simulation study,[8] it was found that $\Gamma_{\iota\xi\kappa\chi}(\omega)$ not only goes to zero at zero frequency, but is a monotonically growing function of ω in the entire domain, such that it has significantly lower values at low ω than in the middle and upper parts of the spectrum, where it reaches its maximum value (cf. Fig. 5 in Ref. 8). Hence, the simulation results of Ref. 8 indicate that the contribution of low-frequency modes to the nonaffine response is small whereas the leading contribution comes from the high-frequency modes. This observation is also in agreement with physical intuition: the source of the nonaffine response is given by the projection of the affine fields on the eigenmodes, which has a higher value the more energetic the modes are.

Based on these observations, one can conclude that the low-frequency modes play a relatively minor role in the nonaffine response as compared to the high-frequency modes. This explains why our theory, which underestimates the low-frequency modes in the case of sphere packings, still yields correct predictions for the shear modulus in excellent agreement with simulations (Fig. 1).

In the case of the bulk modulus, simulations[8] give practically the same behavior for the correlator $\Gamma_{\iota\xi\kappa\chi}(\omega)$ as for the shear modulus, with the low-frequency modes contributing to the nonaffine response to a minor extent. In this case, the failure of the theory in predicting the correct scaling for sphere packings (despite being successful for networks) is more likely to be ascribed to the geometric attenuation of the random affine fields under hydrostatic pressure due to excluded volume, as we speculated in Sec. IV A. However, this hypothesis has to be tested in future work by means of ad hoc numerical studies

as the bulk modulus scaling of packings is a problem currently under debate.[14]

V. CONCLUSION

We have developed an approximate, fully analytical theory of the nonaffine elastic response of amorphous solids that explicitly takes into account the nonaffinity of the atomic displacements. We have applied the nonaffine linear formalism in the formulation of Lemaitre and Maloney[8] to the so-called Cauchy bonded-network model,[1] i.e., to networks of harmonic central-force springs. To evaluate the nonaffine correction to the elastic moduli analytically, an approximation of the Hessian matrix has been proposed where the bond orientation-dependent factors in the Hessian are replaced with their isotropic average (isotropic Hessian). Even though the isotropic Hessian has a density of states that significantly lacks low-frequency modes in comparison with sphere packings, our approximation yields predictions of the shear modulus in excellent quantitative agreement with simulations of sphere packings.[12] The good agreement is explained with the observation, supported by simulations in the literature,[8] that the low-frequency modes, underestimated by our approximation,

play a relatively minor role in the nonaffine response, which is controlled by the upper part of the vibrational spectrum (that is well reproduced by our theory). While our approximation is not suited to accurately describe transport properties of disordered solids in the low-connectivity and low-frequency limits,[18] it seems, on the other hand, successful in accurately describing the elastic response to shear of disordered solids. Furthermore, our theory provides a completely new insight into the linear vanishing of shear rigidity at the isostatic point ($z = 2d$) of disordered solids: this happens because the nonaffine correction at the isostatic point becomes equal in absolute value, but with opposite sign, to the affine part of the shear modulus.

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