Correlations in the Elastic Response of Dense Random Packings

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(Received 2 May 2006; published 20 July 2006)

Results are presented for the autocorrelation function of the vortexlike nonaffine piece of the linear elastic displacement field in dense random bidisperse packings of harmonically repulsive disks in 2D. The autocorrelation function is shown to scale precisely with the length of the simulation cell in systems ranging from 20 to 100 particles across. It is shown that, to first order, the displacement fields can be thought to arise from the action of uncorrelated local random forcing of a homogeneous elastic sheet, and a theory is presented which gives excellent quantitative agreement with the form of the correlation functions. These results suggest measurements to be made in many types of densely packed, random materials where the elastic displacement fields are accessible experimentally such as granular materials, dense emulsions, colloidal suspensions, etc.

DOI: 10.1103/PhysRevLett.97.035503 PACS numbers: 62.20.Dc, 61.43.Fs, 63.50.+x, 81.40.Jj

When Bravais crystals are deformed at zero temperature, the gradient of the displacement field is perfectly homogeneous; that is, the deformation is *affine* [1]. In disordered systems, on the other hand, the response is not affine, even at lowest order in the imposed deformation. These nonaffine deformations are generally observed to lead to reductions in the naive value of the shear modulus which would have been expected based on assumptions of affine deformation. This fact has been noted empirically for many types of disordered systems such as metallic glasses [2–4], emulsions [5–7], granular materials [8–10], and cross-linked, semiflexible polymers [11–13]. In particular, O'Hern and co-workers [14] have shown that these nonaffine corrections to the shear modulus completely dominate the system behavior on approach to the loss of rigidity, the so-called jamming transition [15], during the dilation of repulsive particle packings, and a better understanding of them is of central importance to the larger body of work on the mechanical properties of disordered systems.

Leonforte *et al.,* in a series of works, studied the spatial structure of the nonaffine displacement fields [2–4], which were observed to have a vortexlike structure. They extracted a length scale from the autocorrelation function of these vortexlike fields, noting that the function crossed from positive to negative at a length of about 30 particle diameters, regardless of the size of system being simulated.

More recently, DiDonna and Lubensky (DL) [16] have approached the problem of nonaffine response from within a continuum framework. Their central result was that the autocorrelation of the displacement field decayed like the logarithm of distance (in 2D). Clearly, such logarithmic behavior must somehow be cut off at large distances, but the precise nature of this long range cutoff was not directly addressed by them. Without a more explicit treatment of the large length scales (lengths on the order of the system size), it becomes difficult to determine whether the results of DL are consistent with those of Leonforte *et al.*

Here I follow the basic direction taken by DL; however, the present approach differs in two important ways. First, DL rely on a framework of pairwise interactions within a continuum formalism. The expressions which are derived for the nonaffine displacements are thus necessarily approximate, and it is difficult to ascertain the quality of this approximation. The present approach makes no appeals to coarse-graining, instead directly utilizing the Hessian matrix of the system to derive exact atomistic expressions for the elastic displacement field. Another advantage of focusing directly on the Hessian matrix rather than bond stiffnesses and quenched forces is that it allows one to treat multibody interactions (embedded atom methods, covalent networks, semiflexible polymers, etc.) on the same footing as pairwise interactions, although, *numerically*, we have considered only the latter here. The second, perhaps more crucial, difference is that the present work takes the boundaries of the system explicitly into account, allowing for excellent quantitative agreement with the form of the autocorrelation function which is observed in numerical simulations.

In this Letter, I first briefly review the formalism, presented in detail in Ref. [17], required to compute the linear elastic response of a disordered system. Particular emphasis is placed on the random nature of the strain induced forces $\Xi_{i\alpha}$. I then discuss the numerical protocols used and present results for ensembles of various size. Finally, I show how simple assumptions about the vibrational spectrum and the random nature of $\Xi_{i\alpha}$ can lead to an analytical expression for the autocorrelation function of the displacement field. The theory predicts that the autocorrelation functions for systems of different size should collapse when plotted as a function of distance scaled by the length of the simulation cell L, with the master curve crossing zero at about 0.325.

Throughout this Letter, I use Latin indices to indicate particle identity or to indicate a particular eigenmode of the

system and Greek indices to indicate Cartesian coordinates. Repeated Greek indices should always be summed, while explicit sums will always be written for Latin indices.

Consider a system in a stable mechanical equilibrium state subject to some imposed boundary conditions. Suppose that boundary is deformed affinely. How does the configuration of all the particles change in order to maintain mechanical equilibrium? The answer to this question has long been known, as the same problem must be solved in order to compute the elastic constants in lattices with a basis [18], but it has only more recently been appreciated that the same considerations must apply unaltered to disordered systems [17,19–22]. The fact that the same formalism should apply to a disordered packing should not be surprising; after all, a disordered system (with periodic boundaries) is nothing more than a Bravais lattice with a *huge* number of particles in the basis.

If we first make an affine deformation of the system with $\delta r_{i\alpha} = \gamma y_i \delta_{\alpha x}$, each particle will experience a nonzero force which we denote by $\Xi_{i\alpha}$ and refer to as the *affine forces*. One may compute them directly from the Hessian matrix of the system:

$$\Xi_{i\alpha} \doteq -\frac{\partial F_{i\alpha}}{\partial \gamma} = -\sum_{j\neq i} H_{i\alpha jx} y_{ij}, \tag{1}$$

where y_{ij} is simply $y_i - y_j$. Recall that the Hessian matrix of the system is the second derivative of the energy and first derivative of the forces:

$$H_{i\alpha j\beta} = \frac{\partial^2 U}{\partial r_{i\alpha}\partial r_{j\beta}} = -\frac{\partial F_{i\alpha}}{\partial r_{j\beta}} = -\frac{\partial F_{j\beta}}{\partial r_{i\alpha}}.$$

In order for the system to return to a state of zero force, the corrections to the affine motion $d\mathring{r}_{i\alpha}/d\gamma$ must induce forces which exactly cancel the affine forces:

$$\Xi_{i\alpha} = \sum_j H_{i\alpha j\beta} \frac{d\mathring{r}_{j\beta}}{d\gamma}. \tag{2}$$

The ring over the r denotes the fact that Eq. (2) is an equation for the nonaffine piece of the elastic displacement field. For periodic systems, the solution is defined only up to rigid translations since the Hessian matrix is translationally invariant, and we choose, conventionally, the solution which has no translational component: $\sum_i (d\mathring{r}_{i\alpha}/d\gamma) = 0$.

To study the elastic response of disordered packings, a simple numerical protocol was used. All results are for 2D bidisperse mixutures of harmonically repulsive disks. $U = s^2/2$ for $s > 0$ and 0 otherwise, with $s = 1 - r/(r_a + r_b)$. The mixture is the same as that introduced in related work [17,23,24]. The mixtures used throughout have particles with radii: $r_L = 0.5$ and $r_S = 0.3$ and a number ratio of $N_L = N_S[(1+\sqrt{5})/4]$. The systems are prepared via a zero temperature quench from an initially random state as in Ref. [14]. The initial minimization is performed with a truncated Newton-linear conjugate gradient method using Armijo backtracking with a sufficient decrease parameter of 0.1 [25]. This protocol can be expected to generate configurations which are quite disordered, as no annealing is allowed.

The elements of the Hessian matrix are evaluated using the analytical form for pair potentials. First defining the elementary contributions from individual bonds:

$$M_{i\alpha j\beta} = \left(c_{ij} - \frac{t_{ij}}{r_{ij}}\right) n_{ij\alpha} n_{ij\beta} + \frac{t_{ij}}{r_{ij}} \delta_{\alpha\beta}, \tag{3}$$

where t and c are the first and second derivatives of the bond energy with respect to length, and $n_{ij\alpha}$ is the unit normal pointing from particle i to particle j. Then one obtains the particlewise expression for the Hessian from the individual bond contributions: $H_{i\alpha j\beta} = -M_{i\alpha j\beta}$ for the off diagonal terms ($j \neq i$) and $H_{i\alpha i\beta} = \sum_j M_{i\alpha j\beta}$ for the diagonal terms ($j = i$). In order to compute the nonaffine response, the field $\Xi_{i\alpha}$ is first evaluated using (1) and (3), then (2) is solved via a linear conjugate gradient routine; note that explicit finite strain and energy reminimization is only ever imposed on the system as a check on the derivation of Eq. (2).

In Fig. 1, we show the fields Ξ and $\frac{d\mathring{r}}{d\gamma}$ for a typical 30×30 system. As expected, based on the fact that these systems are not allowed to anneal at all, Ξ appears completely random. The spatial autocorrelation function of the Ξ field decays to zero within 2 particle radii (not shown). The nonaffine displacement, on the other hand, shows the striking "vortexlike" features which have been observed earlier by others [2–4,16].

To study such vortexlike displacement fields, Tanguy *et al.* [3] have suggested using the standard 2 point autocorrelation function (introducing the notation $v_{i\alpha} \doteq d\mathring{r}_{i\alpha}/d\gamma$):

$$g(\vec{\delta}) \doteq \int v_\alpha(\vec{r} + \vec{\delta}) v_\alpha(\vec{r}) d\vec{r}. \tag{4}$$

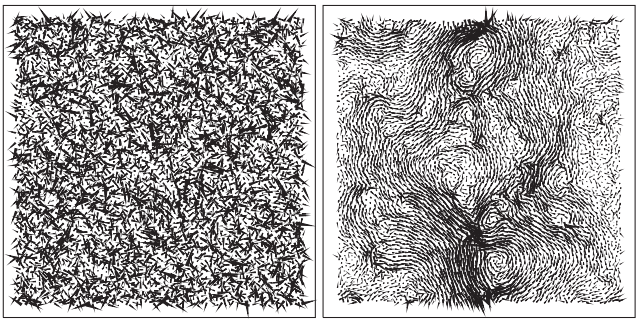

FIG. 1. Reprinted from Ref. [17] with the kind permission of Springer Science and Business Media. The fields $\Xi_{i\alpha}$ (left) and $d\mathring{r}_{i\alpha}/d\gamma$ (right) for a typical 30×30 system. Note the random nature of Ξ in contrast to the striking correlations in $\frac{d\mathring{r}}{d\gamma}$.

One often then takes averages over angles to define a 1D $g(\delta)$. The interest of Tanguy *et al.* in $g(\delta)$ is that, while at small δ, the correlation function must be positive; when δ becomes larger than some size which characterizes the vortices, g should cross from positive to negative. The characteristic length can then be identified with the zero crossing of $g(\delta)$. We plot this autocorrelation function for our system in Fig. 2.

To compute an approximate analytical form for $g(\vec{\delta})$, first expand (2) in terms of the eigenmodes of the Hessian to get

$$v_{i\alpha} = \sum_p \frac{\psi^p_{i\alpha}}{(\lambda_p/\Xi_p)},$$

where $\psi^p_{i\alpha}$ is the pth eigenmode of the Hessian and Ξ_p is the projection of $\Xi_{i\alpha}$ onto the pth eigenmode: $\Xi_p = \sum_i \Xi_{i\alpha}\psi^p_{i\alpha}$. First note that if the field $\Xi_{i\alpha}$ is random, it should be random in any basis, so we may safely suppose that the Ξ_p is a random Gaussian variable. Next, in analogy with DL, we suppose that the eigenmodes of the Hessian can be approximated (to first order) as the transverse and longitudinal plane waves which are the eigenstates of the Lamé-Navier operator [26].

$$v_{i\alpha L(T)} = \tilde{\mu}^{-1}_{L(T)} \sum_{m,n} \frac{\hat{p}^{L(T)}_{mn\alpha}}{1/(\Xi_{mn})} \frac{e^{i2\pi(mx_i+ny_i)/L}}{\sqrt{m^2+n^2}} \qquad (5)$$

[27]. Here $\hat{p}^{L(T)}_{mn\alpha}$ is the polarization vector for the longitudinal (transverse) wave and $\tilde{\mu}_{L(T)}$ is an effective renormalized stiffness (which is *not* discussed further here) for the longitudinal (transverse) waves. This expression explains the fact that Leonforte *et al.* observe amplitudes proportional to $1/k$ in the Fourier decomposition (both longitudinal and transverse) of their displacement fields [4].

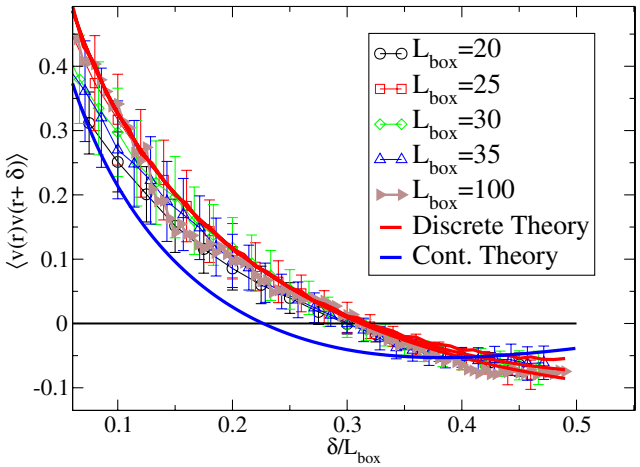

FIG. 2 (color online). $g(\delta/L)/g(0)$ for systems of various L, where L is the cell length. The 4 smallest sizes of 20×20, 25×25, 30×30, and 35×35 represent data averaged over ensembles of 16 assemblies at each size. The data for the 100×100 box correspond to a single system. Theoretically predicted curves are superimposed on the data. The dark (blue) curve is obtained by the semicontinuous approximation $\int_1^\infty (2\pi/k)J_0(2\pi k\delta)dk$, while the 3 light (red) curves are partial sums over the first 40 terms in (6) with the delta vector lying at angles of 0, $\pi/8$, and $\pi/4$.

To compute the autocorrelation function, one simply uses the convolution theorem; since waves with different wave vector or polarization are uncorrelated, the only contributions come from individual waves:

$$g(\vec{\delta}) = \left(\frac{\langle\Xi^2\rangle}{\tilde{\mu}^2_L} + \frac{\langle\Xi^2\rangle}{\tilde{\mu}^2_T}\right)\sum_{mn} \frac{\cos(2\pi(m\delta_x + n\delta_y)/L)}{m^2+n^2}. \qquad (6)$$

Technically, the sum should have a Debye-like cutoff at large m, n to account for the finite number of eigenmodes in the finite system, but we neglect this cutoff here as we will only be numerically evaluating finite partial sums. One often approximates such sums by an isotropic integral over $\vec{k}$ which reduces to an integral of the Bessel function J_0, $\int_1^\infty (2\pi/k)J_0(2\pi k\delta)dk$; however, such an approach is bound to fail when δ is of the same order as the length of the box. This should be obvious, as one must have the translational symmetry required of g; namely, $g(x, y) = g(x + mL, y + nL)$. The square shape of the cell also dictates that g should, strictly speaking, be *anisotropic*. The curve corresponding to a horizontal separation must, by symmetry, be flat as it approaches 0.5, while the curve corresponding to a diagonal separation becomes flat only at $0.5\sqrt{2}$ and is still decreasing at 0.5.

Figure 2 shows the numerically obtained $g(\delta)$ curves along with partial sums of (6) (taken for 3 different angles of $\vec{\delta}$: 0, $\pi/8$, and $\pi/4$) and the integral over J_0. Theoretical curves are scaled arbitrarily, but this single fitting parameter does not affect, e.g., the location of the zero crossing. The first 40 terms of the sum are used and we find that the curve is well converged in the range plotted, with slower convergence at short δ due to the coherence of successive modes for small δ. Note that our data are not inconsistent with the main conclusion of DL, that the correlation function should decay as the log of the distance up to the system size cutoff, and that we plot the data in this way in Fig. 3. However, the limits in which DL evaluate their integral analogs of the sum (6) to obtain expressions for $g(\delta)$ in real space make it difficult to compute quantities such as the zero crossing point, while the fully discrete theory has no problems.

To summarize, we have shown that, to first order in the disorder, the nonaffine linear displacements of our random 2D packings can be thought of as the displacement fields generated by locally random forces applied to a *homogeneous* elastic sheet. As the elastic sheet possesses no characteristic length, the only length which emerges is the length of the box itself.

One outstanding problem is that Refs. [2–4] show a well defined characteristic length, in apparent contradiction with our results. I can identify essentially two possible resolutions. First, it is possible that the eigenmodes of the

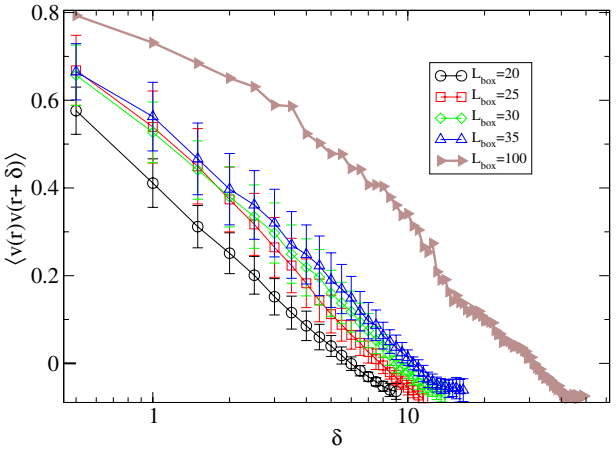

FIG. 3 (color online). Data from Fig. 2, plotted as a function of $\ln(\delta)$.

Hessian are not, in fact, well approximated by plane waves. Silbert and co-workers [28] have pointed out that, near the onset of rigidity loss in repulsive systems, the approximation of the eigenmodes by plane waves does indeed become bad. However, it seems unlikely that this is the case for the systems in Ref. [3], as this approximation was checked directly and found to be reasonably good.

A more likely explanation is the annealing protocol to which the systems in Refs. [2–4] are subjected. The systems here are subjected to the most violent quench protocol conceivable, whereas those in Refs. [2–4] have been well annealed. It would be interesting, indeed, to measure the local Ξ field as prepared in Refs. [2–4] to rule out the lack of local spatial correlations which were assumed in the derivation here. Such correlations could provide an intrinsic length scale and might explain the lack of scaling of $g(\delta)$ with the system size; however, one would expect to regain this scaling eventually for large enough samples. It would be straightforward to incorporate these correlations in the expression for the correlation function.

This material is based upon work supported by the National Science Foundation under Grants No. DMR 0454947 and No. PHY99-07949. I thank A. Lemaître and M. O. Robbins for many useful discussions.

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