Packing of Compressible Granular Materials

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(Received 25 August 1999; revised manuscript received 22 December 1999)

3D computer simulations and experiments are employed to study random packings of compressible spherical grains under external confining stress. In the rigid ball limit, we find a continuous transition in which the stress vanishes as $(\phi - \phi_c)^\beta$, where ϕ is the (solid phase) volume density. The value of ϕ_c depends on whether the grains interact via only normal forces (giving rise to random close packings) or by a combination of normal and friction generated transverse forces (producing random loose packings). In both cases, near the transition, the system's response is controlled by localized force chains.

PACS numbers: 81.05.Rm

Dense packings of spherical particles are an important starting point for the study of simple liquids, metallic glasses, colloids, biological systems, and granular matter [1–4]. In the case of liquids and glasses, finite temperature molecular dynamics studies of hard sphere models have been particularly important. Here one finds a first order liquid-solid phase transition as the solid phase volume fraction, ϕ, increases. Above the freezing point, a metastable disordered state can persist until $\phi \to \phi_{\rm RCP}^-$ [4], where $\phi_{\rm RCP}$ is the density of random close packing (RCP)—the densest possible random packing of hard spheres. This Letter is concerned with the nonlinear elastic properties of granular packings. Unlike glasses and amorphous solids, this is a zero temperature system in which the interparticle contact forces are nonlinear, purely repulsive, and path (i.e., history) dependent.

In the conventional continuum approach to this problem, the granular material is treated as an elasto-plastic medium [5]. However, this approach has been challenged by recent authors [6] who argue that granular packings represent a new kind of *fragile* matter and that more exotic methods, e.g., the fixed principal axis ansatz, are required to describe their internal stress distributions. These new continuum methods are complemented by microscopic studies based on either contact dynamics simulations of *rigid* spheres or statistical models, such as the q model, which does not take explicit account of the character of the intergrain forces [7,8].

In our view, a proper description of the stress state in granular systems must deal with the fact that the individual grains are deformable. We report here on a 3D study of deformable spheres interacting via Hertz-Mindlin contact forces [9]. Our simulations cover four decades in the applied pressure and allow us to understand the regimes in which the different theoretical approaches described above are valid. Since the grains in our simulations can deform, the volume fraction can be increased above the hard sphere limit and we are able to study the approach to the RCP and random loose packed (RLP) [3] states from this realistic perspective. Within this framework, the fragile rigid grain limit is described as a continuous phase transition where the applied stress, σ, vanishes continuously as $(\phi - \phi_c)^\beta$. Here ϕ_c is the critical volume density, and β is the corresponding critical exponent.

Of particular importance is the fact that ϕ_c depends on the type of interaction between the grains. If they interact via normal forces only [10], they slide and rotate freely mimicking the rearrangements of grains during shaking in experiments [1,2]. We then obtain the RCP value $\phi_c = 0.634(4)\,(\approx \phi_{\rm RCP})$. By contrast, if the grains interact by combined normal and friction generated transverse forces, we get RLP at the critical point with $\phi_c = 0.6284(2) < \phi_{\rm RCP}$. Our results indicate that the transitions at both RCP or RLP are driven by localized force chains. Concomitant with the approach to the critical volume fraction we find a change in the probability distribution and in the degree of spatial correlation of the interparticle forces.

Numerical simulations.—To understand better the behavior of real granular materials, we perform granular dynamics simulations of unconsolidated packings of deformable spherical glass beads using the discrete element method developed by Cundall and Strack [11]. Unlike previous work on rigid grains, we work with a system of deformable elastic grains interacting via normal and tangential Hertz-Mindlin forces plus viscous dissipative forces [9]. The grains have shear modulus 29 GPa, Poisson's ratio 0.2, and radius 0.1 $\pm$ 0.005 mm.

Our simulations employ periodic boundary conditions and begin with a gas of 10 000 nonoverlapping spheres located at random positions in a cube 4 mm on a side. Generating a mechanically stable packing is not a trivial task [4]. At the outset, a series of strain-controlled isotropic compressions and expansions are applied until a volume fraction slightly below the critical density is reached. At this point the system is at zero pressure and zero coordination number. We then compress along the z direction, until the system equilibrates at a desired vertical stress σ and a nonzero average coordination number $\langle Z \rangle$.

Figure 1a shows the behavior of the stress as a function of the volume fraction. We find that the pressure vanishes at a critical $\phi_c = 0.6284(2)$. Although we cannot rule out a discontinuity in the pressure at ϕ_c—as we could expect

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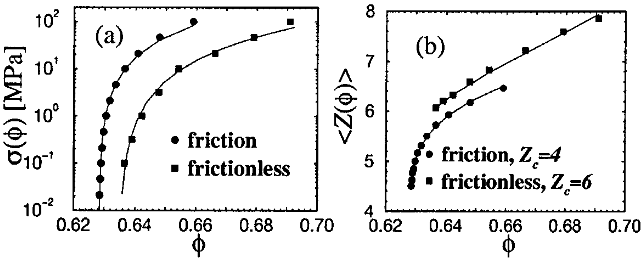

FIG. 1. (a) Confining stress and (b) average coordination number as a function of volume fraction for friction and frictionless balls.

for a system of hard spheres—our results indicate that the transition is continuous and the behavior of the pressure can be fitted to a power law form

$$\sigma \sim (\phi - \phi_c)^{\beta}, \tag{1}$$

where $\beta = 1.6(2)$. Our 3D results contrast with recent experiments of slowly sheared grains in 2D Couette geometries [12] where a faster than exponential approach to ϕ_c was found, while they agree qualitatively with similar continuous transitions found in compressed emulsions and foams [10].

Figure 1b shows the behavior of the mean coordination number, $\langle Z\rangle$, as a function of ϕ. We find

$$\langle Z\rangle - Z_c \sim (\phi - \phi_c)^{\theta}, \tag{2}$$

where $Z_c = 4$ is a minimal coordination number, and $\theta = 0.29(5)$ is a critical exponent. At criticality the system is very loose and fragile with a very low coordination number. The value of Z_c can be understood in terms of constraint arguments as discussed in [13]; in the rigid ball limit, for a disordered system with both normal and transverse forces, we find $Z_c = D + 1 = 4$ [13]. As we compress the system more contacts are created, providing more constraints so that the system becomes overdetermined.

We notice that ϕ_c obtained for this system is considerably lower than the best estimated value at RCP, $\phi_{\rm RCP} = 0.6366(4)$ obtained by Finney [2] using ball bearings. This latter value is obtained by carefully vibrating the system and letting the grains settle into the most compact packing. Numerically, this is achieved by allowing the grains to reach the state of mechanical equilibrium interacting only via normal forces, since frictionless grains can slide freely and find more compact packings than friction grains. Numerically we confirm this by equilibrating the system at zero transverse force. The critical packing fraction found in this way is $\phi_c = 0.634(4)$ ($\approx \phi_{\rm RCP}$ within error bars). The stress behaves as in Eq. (1) but with a different exponent $\beta = 2.0(2)$ (Fig. 1a). At the critical volume fraction the average coordination number is now $Z_c = 6$ [and $\theta = 0.94(5)$, Fig. 1b], which again can be understood using constraint arguments which would give a minimal coordination number $Z_c = 2D$ for frictionless rigid balls [13].

We conclude that the value $\phi_c \approx 0.6284 < \phi_{\rm RCP}$ found with transverse forces corresponds to the RLP limit, experimentally achieved by pouring balls in a container but without allowing for further rearrangements [3]. Experimentally, stable loose packings of repulsive hard spheres with ϕ as low as 0.60 have been found [3]. In our simulations, ϕ_c lower than 0.6284 can be obtained by increasing the strength of the tangential forces. This is in agreement with experiments of Scott and Kilgour [14] who found that the maximum packing density of spheres decreases as the surface roughness (friction) of the balls increases.

While previous studies characterized RCP's and RLP's by using radial distribution functions and Voronoi constructions [2], we take a different approach which allows us to compare our results directly with recent work on force transmissions in granular matter. Previous studies of granular media indicate that, for forces greater than the average value, the distribution of intergrain contact forces is exponential [7,8]. In addition, photoelastic visualization experiments and simulations [7,15] show that contact forces are strongly localized along "force chains" which carry most of the applied stress. The existence of force chains and exponential force distributions are thought to be intimately related.

Here we analyze this scenario in the entire range of pressures: from the ϕ_c limit and up. Figure 2a shows the force distribution obtained in the simulations with friction balls. At low stress, the distribution is exponential in agreement with previous experiments and models. However, when the system is compressed further, we find a gradual transition to a Gaussian force distribution. We observe a similar transition (not shown here) in our simulations involving frictionless grains under isotropic compression. This suggests that our results are generic and do not depend,

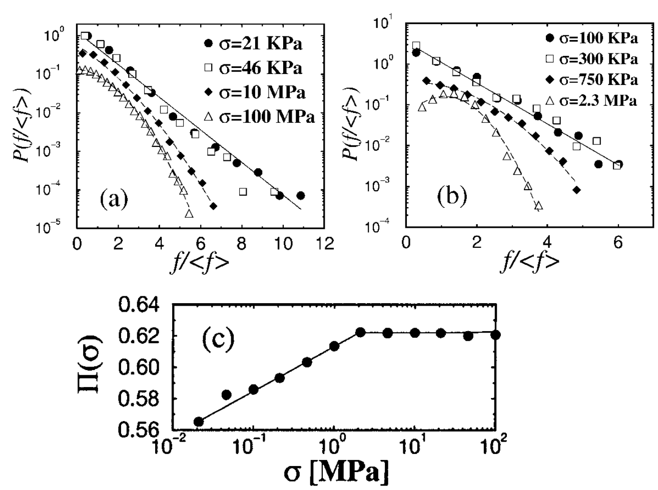

FIG. 2. Distribution of forces for different confining stresses σ obtained (a) in the numerical simulations of friction balls and (b) in the carbon paper experiments. The straight solid lines are fittings to exponential forms and the dashed lines are fittings to Gaussian forms. In both graphs we shift down the distributions corresponding to the two larger stresses for clarity. (c) Participation number versus external stress for the same system as in (a).

qualitatively, on the preparation history or on the existence of friction generated transverse forces between the grains.

Physically, we find that the transition from Gaussian to exponential force distribution is driven by the localization of force chains as the applied stress is decreased. In granular materials, with particles of similar size, localization is induced by the disorder of the packing arrangement. To quantify the degree of localization, we consider the participation number Π:

$$\Pi \equiv \left(M \sum_{i=1}^{M} q_i^2 \right)^{-1}. \tag{3}$$

Here M is the number of contacts between the spheres, $\langle Z \rangle = 2M/N$, and N is the number of spheres. $q_i \equiv f_i / \sum_{j=1}^{M} f_j$, where f_i is the magnitude of the total force at every contact. From the definition (3), $\Pi = 1$ indicates a limiting state with a spatially homogeneous force distribution ($q_i = 1/M, \forall\ i$). On the other hand, in the limit of complete localization, $\Pi \approx 1/M \rightarrow 0$ as $M \rightarrow \infty$.

Figure 2c shows our results for Π versus σ. Clearly, the system is more localized at low stress than at high stress. Initially, the growth of Π is logarithmic, indicating a smooth delocalization transition. This behavior is seen up to $\sigma \approx 2.1$ MPa, after which the participation number saturates to a higher value:

$$\begin{aligned} \Pi(\sigma) &\propto \log(\sigma) \qquad (\sigma < 2.1\ \text{MPa}), \\ \Pi(\sigma) &\approx 0.62 \qquad (\sigma > 2.1\ \text{MPa}). \end{aligned} \tag{4}$$

This behavior suggests that, near the critical density, the forces are localized in force chains sparsely distributed in space. As the applied stress is increased, the force chains become more dense and are thus distributed more homogeneously.

How might we expect the participation number to depend upon other system parameters when the forces are transmitted principally by force chains? In an idealized situation, the system has N_{FC} force chains, each of which has N_z spheres. Each sphere in a force chain has two major load bearing contacts, which loads must be approximately equal. In the lateral directions, roughly four weak contacts are required for stability. These contacts carry a fraction $\alpha < 1$ of the major vertical load. All other contacts have $f_i \approx 0$. Under these assumptions,

$$\Pi = \frac{2}{\langle Z \rangle} \frac{(1 + 2\alpha)^2}{(1 + 2\alpha^2)} \frac{N_{FC} N_z}{N} \leq \frac{2}{\langle Z \rangle} \frac{(1 + 2\alpha)^2}{(1 + 2\alpha^2)}. \tag{5}$$

The last inequality becomes an equality if and only if all the balls are in force chains. From our simulations at large pressure $\alpha \approx 2/5$, so at $\langle Z \rangle \approx 8$, $\Pi \approx 0.62$, which implies that the system has been completely homogenized. Although Eq. (5) is oversimplified, we believe that the change in slope in Fig. 2c is emblematic of the complete disappearance of well-separated chains.

The localization transition can be understood by studying the behavior of the forces during the loading of the sample. Clearly, visualizing forces in 3D systems is a complicated task. In order to exhibit the rigid structure from the system we visually examine all the forces larger than the average force; these carry most of the stress of the system [8,15]. We look for force chains by starting from a sphere at the top of the system and following the path of maximum contact force at every grain. We look only for the paths which percolate, i.e., stress paths spanning the sample from the top to the bottom. In Fig. 3 we show the evolution of the force chains thus obtained for two extreme cases of confining stress. We clearly see localization at low confining stress: the force-bearing network is concentrated in a few percolating chains. At this point the grains are weakly deformed but still well connected. We expect a broad force distribution, as found in this and previous studies. As we compress further, new contacts are created and the density of force chains increases. This in turn gives rise to a more homogeneous spatial distribution of forces, which is consistent with the crossover to a narrow Gaussian distribution.

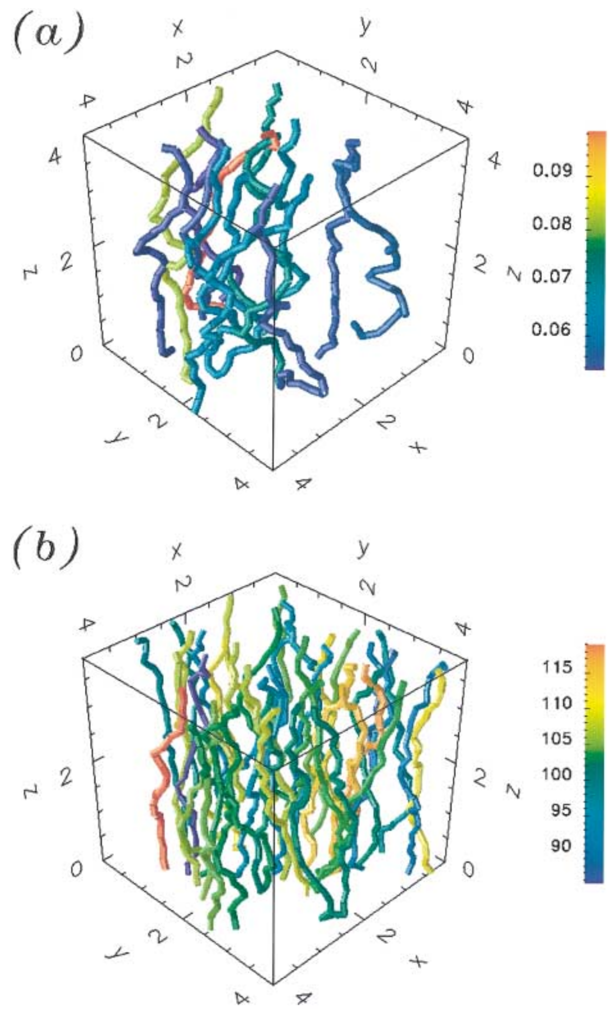

FIG. 3 (color). Example of percolating force chains for the same system as in Fig. 2a: (a) near ϕ_c ($\sigma = 21$ kPa); (b) away from ϕ_c ($\sigma = 100$ MPa). The color code is according to the total force, in N, carried by the chains.

Experiments.—Some of the predictions of our numerical study can be tested using standard carbon paper experiments [7], which have been used successfully in the past to study the force fluctuations in granular packings. A Plexiglas cylinder, 5 cm diameter and varying height (from 3 to 5 cm), is filled with spherical glass beads of diameter 0.8 ± 0.05 mm. At the bottom of the container we place a carbon paper with white glossy paper underneath. We close the system with two pistons and we allow the top piston to slide freely in the vertical direction, while the bottom piston is held fixed to the cylinder. The system is compressed in the vertical direction with an Inktron™ press and the beads at the bottom of the cylinder left marks on the glossy paper. We digitize this pattern and calculate the "darkness" [7] of every mark on the paper. To calibrate the relationship between the marks and the force, a known weight is placed on top of a single bead; for the forces of interest in this study (i.e., from ≈0.05 to 6 N), there is a roughly linear relation between the darkness of the dot and the force on the bead.

We perform the experiment for different external forces, ranging from 2000 to 9000 N, and different cylinder heights. The corresponding vertical stress, σ, at the bottom of the cylinder ranges between 100 kPa and 2.3 MPa (as measured from the darkness of the dots). The results of four different measurements are shown in Fig. 2b. For σ smaller than ≈750 kPa, the distribution of forces, f, at the bottom piston decays exponentially:

$$P(f) = \langle f \rangle^{-1} \exp[-f/\langle f \rangle], \quad (\sigma < 750 \text{ kPa}), \quad (6)$$

where $\langle f \rangle$ is the average force. When the stress is increased above 750 kPa there is a gradual crossover to a Gaussian force distribution as we find in the simulations. For example, at 2.3 MPa we have

$$P(f) \propto \exp[-k^2(f - f_o)^2], \quad [\sigma = 2.3 \text{ MPa}], \quad (7)$$

where $kf_o \approx 1$, and therefore $\langle f \rangle \approx f_o$. Similar results have been found in 2D geometries [12].

Conclusion.—In summary, using both numerical simulations and experiments, we have studied compressible granular media in a range of pressures spanning almost four decades. In the limit of weak compression, the stress vanishes continuously as $(\phi - \phi_c)^\beta$, where ϕ_c corresponds to RLP or RCP according to the existence or not of transverse forces between the grains, respectively. At criticality, the coordination number approaches a minimal value Z_c (equals 4 for friction and equals 6 for frictionless grains) also as a power law. Our result $Z_c = 6$ agrees with experimental analysis of Bernal packings for close contacts between spheres fixed by means of wax [1], and our own analysis of the Finney packings [2] using the actual sphere center coordinates of 7934 steel balls. However, no similar experimental study exists for RLP which could be able to confirm $Z_c = 4$. A critical slowing down—the time to equilibrate the system increases near ϕ_c—and the emergence of shear rigidity (to be discussed elsewhere) is also found at criticality. The distribution of forces is found to decay exponentially. The system is dominated by a fragile network of relatively few force chains which span the system.

When the stress is increased away from ϕ_c to the point that the number of contacts has significantly increased from its initial value Z_c, we find the distribution of forces crosses over to a Gaussian and the participation number increases and then abruptly saturates. The system has become elastic and homogeneous down to a scale comparable to the grain size. Our simulations indicate that the crossover is associated with a loss of localization and the ensuing homogenization of the force-bearing stress paths.

We thank B. Andrews, J. St. Germain, N. Gland, and D. Pissarenko for help and discussions.

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