

Entropic origin of stress correlations in granular materials

G. Lois^{1,2}, J. Zhang³, T. S. Majmudar³, S. Henkes⁴, B. Chakraborty⁴, C. S. O'Hern^{1,2}, and R. P. Behringer³

¹Department of Mechanical Engineering, Yale University, New Haven, Connecticut 06520-8284

²Department of Physics, Yale University, New Haven, Connecticut 06520-8120, USA

³Department of Physics, Duke University, Box 90305, Durham, NC 27708, USA

⁴Department of Physics, Brandeis University, Waltham, MA 02454, USA

(Dated: October 26, 2018)

We study the response of granular materials to external stress using experiment, simulation, and theory. We derive an entropic, Ginzburg-Landau functional that enforces mechanical stability and positivity of contact forces. In this framework, the elastic moduli depend only on the applied stress. A combination of this feature and the positivity constraint leads to stress correlations whose shape and magnitude are extremely sensitive to the applied stress. The predictions from the theory describe the stress correlations for both simulations and experiments semiquantitatively.

A striking feature of dry granular materials and other athermal systems is that they form force chain networks in response to applied stress, such that large forces are distributed inhomogeneously into linear chain-like structures [1, 2]. A number of experimental studies have visualized and quantified these networks in granular systems using carbon paper [3] and photoelastic techniques [4, 5]. These studies demonstrated that geometrical and mechanical properties of force chain networks are acutely sensitive to preparation procedures, especially near the jamming transition[6]. For example, in isotropically compressed systems, force networks are ramified with only short-ranged spatial correlations of the stress. In contrast, in sheared systems, aligned force chains give rise to longer ranged stress correlations in the compressive direction. Since granular (or other) systems near jamming are fragile and highly sensitive to preparation, one expects that their mechanical properties near jamming might not be captured by simple linear elastic response [7].

Developing alternative theoretical descriptions for granular media is challenging for several important reasons [8, 9, 10, 11]: (1) since tensile stresses are absent in dry granular materials they only remain intact via applied stress, making the limiting zero-stress isostatic state, where the number of degrees of freedom matches the number of constraints[12, 13, 14], problematic; (2) forces at the microscopic level are indeterminant due to friction and disorder; (3) granular materials are athermal, so that conventional energy-based statistical approaches are not appropriate; and (4) near isostaticity we expect fluctuations to be important, both within a single realization of a system and from realization to realization. New methods are needed to bridge the gap between force networks at small length scales and continuum elasto-plastic theory at large scales, and to capture the highly sensitive, fluctuating behavior of granular systems near jamming.

We construct a model for stress fluctuations based on grain-scale force and torque balance and positivity of contact forces rather than energy conservation. We then calculate stress correlations and predict differences for

systems under isotropic compression versus shear stress. We also perform complementary numerical simulations and experimental studies of jammed granular systems in 2D subject to isotropic compression and pure shear. The stress correlation functions from theory, simulation, and experiment are in qualitative and in some cases quantitative agreement. In particular, the theory predicts that the form of the stress correlations depends on how the jammed states were prepared.

Theoretical Framework: Fluctuations are inherently related to the number of microscopic states available under a given set of macroscopic conditions. In equilibrium thermodynamics, the microcanonical entropy describes the nature of fluctuations and response. When we turn to granular systems, the identification of states by energy is no longer useful, and a different criterion for identifying states is needed. The approach that we pursue here exploits a different conservation principal, based on force and torque balance, which applies rigorously for granular materials [15, 16, 17].

The force-moment tensor of mechanically stable (MS) packings, $\hat{\Sigma} = \int d^d r \hat{\sigma}(\mathbf{r})$, where $\hat{\sigma}(\mathbf{r})$ denotes the local stress tensor, is an extensive variable that is a topological invariant [15, 20]. In the force-moment ensemble, $\hat{\Sigma}$ remains fixed barring system-spanning changes in $\hat{\sigma}$. Hence local fluctuations and response only involve grain configurations with the same $\hat{\Sigma}$. To construct a theory for stress correlations, we adopt a coarse-grained approach, in which jammed configurations are represented by a continuous field [21] and the entropy $S(\hat{\Sigma})$ is defined via an appropriate Ginzburg-Landau functional. The theory should be valid close to the jamming transition where grains have negligible deformations, and stress fluctuations decouple from volume fluctuations. The decoupling is exact only in the limit of infinitely rigid grains [18].

In both two and three dimensions, a continuous field can be defined that upholds force and torque balance [19, 20] of granular packings. We will focus on 2D systems, where a *scalar* field Ψ , the Airy stress function [7], is

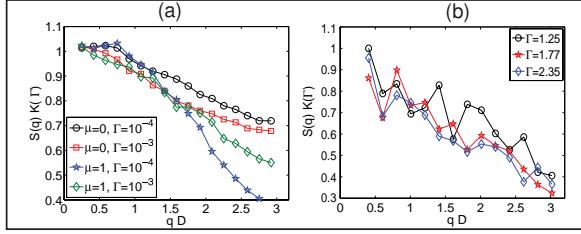


FIG. 1: Pressure correlations in Fourier space for pure compression. Angle-averaged $S(q)K(\Gamma)$ from (a) simulations at different Γ (reduced units) and static friction coefficient μ , (b) experiments at different Γ (in units of $N \cdot m$), $\mu = 0.7$ and z ranging from 3.35–3.68. The experimental data have been scaled by $K(\Gamma) = (z_{\text{iso}}/2 + c(z - z_{\text{iso}}^2))/\Gamma^2$ [20], with $c = 2.8$. Theory predicts that $S(q)K(\Gamma)$ is independent of Γ for $q \ll 1/\xi$.

related to the local stress tensor by

$$\hat{\sigma}(\mathbf{r}) = \begin{bmatrix} \partial_y^2 \Psi & -\partial_x \partial_y \Psi \\ -\partial_x \partial_y \Psi & \partial_x^2 \Psi \end{bmatrix}. \quad (1)$$

We define $\Gamma = \text{Tr}\hat{\Sigma}$ and $\tau = s_1 - s_2 = \sqrt{\Gamma^2 - 4(\det \Sigma)^2}$, where $s_1 > s_2$ are the eigenvalues of $\hat{\Sigma}$. Given a $\hat{\Sigma}$, Ψ can be expanded as $\Psi_0 + \psi$, where ψ represents fluctuations around Ψ_0 . The field Ψ_0 satisfies the biharmonic equation, $\nabla^4 \Psi_0 = 0$ [7], and $\Sigma_{ij} = \int d^d r (\delta_{ij} \nabla^2 \Psi_0 - \partial_i \partial_j \Psi_0)$ [20].

The probability for fluctuations ψ can be written as $P[\psi] = Z^{-1}(\hat{\Sigma}) e^{-L[\Psi_0, \psi]} \equiv Z^{-1}(\hat{\Sigma}) e^{-L_{\hat{\Sigma}}[\psi]}$. The functional $L_{\hat{\Sigma}}[\psi]$ measures the contribution to the entropy from all grain packings that have a coarse-grained representation $\psi(r)$. The partition function $Z(\hat{\Sigma}) \equiv e^{S(\hat{\Sigma})} = \int D\psi e^{-L_{\hat{\Sigma}}[\psi]}$ generates correlators of the field ψ [21]. Because of gauge freedom, $L_{\hat{\Sigma}}[\psi]$ can only depend on second derivatives of ψ . Independent second order scalars involving second derivatives of ψ can be constructed from the invariants of the local stress tensor, $\text{Tr}(\hat{\sigma})$ and $\det(\hat{\sigma})$. The coefficients of these terms and, therefore, the nature and strength of fluctuations are controlled by the field Ψ_0 (or $\hat{\Sigma}$). To lowest order in ψ , $L_{\hat{\Sigma}}[\psi]$ resembles the free energy for an elastic material in two dimensions [7]:

$$L_{\hat{\Sigma}}[\psi] = \int d^2 r \left\{ \alpha_1(\hat{\Sigma}) (\partial_x^2 \psi)^2 + \alpha_2(\hat{\Sigma}) (\partial_y^2 \psi)^2 + \alpha_3(\hat{\Sigma}) (\partial_x \partial_y \psi)^2 + \alpha_4(\hat{\Sigma}) (\partial_x^2 \psi)(\partial_y^2 \psi) \right\}. \quad (2)$$

The crucial differences between the description in Eq. 2 and traditional elasticity theory are (a) the stiffness constants are determined by Ψ_0 or $\hat{\Sigma}$, and thus the theory is inherently nonlinear, and (b) the origin of $L_{\hat{\Sigma}}[\psi]$ is entropic. The functional $L_{\hat{\Sigma}}[\psi]$ can be used to calculate averages and correlation functions. Below, we consider the behavior of pressure correlation functions under isotropic compression and pure shear using Eq. (2).

Isotropic Compression: For isotropic compression with no deviatoric stress, the stiffness constants only depend on Γ , and $L_{\Gamma}[\psi]$ is isotropic:

$$L_{\Gamma}[\psi] = \int d^2 r \left\{ \frac{K(\Gamma)}{2} \{(\partial_x^2 \psi)^2 + (\partial_y^2 \psi)^2\} + \lambda(\Gamma) (\partial_x \partial_y \psi)^2 + (K(\Gamma) - \lambda(\Gamma)) \partial_x^2 \psi \partial_y^2 \psi \right\}, \quad (3)$$

with two stiffness constants K and λ . The positivity of contact forces implies that both $\partial_x^2 \Psi$ and $\partial_y^2 \Psi$ must be non-negative [15, 20]. This is a difficult constraint to impose exactly on the fluctuating field ψ . However, it can be enforced in a mean-field way by requiring that $K(\Gamma) \geq 1/\Gamma^2$, which guarantees that the amplitude of the long-wavelength fluctuations are such that the positivity criterion is met [20, 22]. The inequality constraint on $K(\Gamma)$ implies that different preparation histories can lead to different fluctuations. The maximum entropy of jammed packings is achieved in protocols that meet the equality, and we focus here on these marginal packings. Note that the positivity constraint does not impose any conditions on λ , which is therefore taken to be independent of Γ . Near jamming when $\Gamma \rightarrow 0$, $K(\Gamma)$ becomes arbitrarily large and the λ terms can be ignored [23].

The results for the correlations of the local pressure are best visualized in Fourier space. From Eq. (3), these correlations are predicted to be isotropic:

$$S(\mathbf{q}) = \langle |\delta\Gamma(\mathbf{q})|^2 \rangle = q^4 \langle |\psi(\mathbf{q})|^2 \rangle = \frac{K^{-1}(\Gamma)}{1 + \xi^2 q^2}, \quad (4)$$

where $\delta\Gamma = \nabla^2 \psi$, \mathbf{q} is the wavevector, and ξ is a correlation length that describes the decay of correlations at large q , and is defined by higher order terms not included in Eq. (3). In an experiment or simulation at fixed Γ/A , there are many MS packings, and each is characterized by a continuously varying field $\psi(\mathbf{r})$. The spatial correlations of stress, for a given Γ , are determined by averaging over these configurations. If the configurations are sampled according to the theoretically predicted $P[\psi] \propto e^{-L_{\Gamma}[\psi]}$, the correlations measured in simulations and experiments should be well described by the field-theoretic predictions. Since frictionless granular packings are isostatic near jamming [24], an exact calculation yields $K(\Gamma) = z_{\text{iso}}/(2\Gamma^2)$ [20], where the number of contacts $z_{\text{iso}} = 4$ in 2D. In contrast, frictional packings have $z_{\text{iso}} = 3$ in 2D, but are often hypostatic [25]. We do not have an exact result for $K(\Gamma)$ away from isostaticity, however, a form that has been successful [20] is $K(\Gamma) = (z_{\text{iso}}/2 + c(z - z_{\text{iso}}^2))/\Gamma^2$, where c is a phenomenological constant. We will use this form to compare the predictions with results from frictional packings.

To test the q -space pressure fluctuations predicted in Eq. (4), we have numerically generated MS packings of bidisperse disks ($N/2$ large and $N/2$ small particles

with diameter ratio $r = 1.4$) both with and without friction near the jamming transition using well-known packing-generation algorithms [26, 27]. For frictionless grains, these algorithms generate packings at the margin of stability [14]. We studied system sizes ranging from $N = 256$ to 4096 , systems with square cells and periodic boundary conditions, pressures in the range $\Gamma/A = 10^{-5}$ to 10^{-3} (in reduced units of the grain stiffness), and static friction coefficients in the range $\mu = [0, 1]$. We have also carried out experiments using a biaxial apparatus that has been described previously [6, 28, 29]. The biax is a device that allows us to apply highly controlled deformations to quasi-2D systems of photoelastic disks. By using photoelastic disks, it is possible to obtain all contacts and contact forces in the system. In this study, contact forces are calculated for $N = 1228$ disks, with $N/5$ large and $4N/5$ small disks with diameter ratio $r = 1.2$ and coefficient of static friction $\mu = 0.7$. The experimental protocol generates packings farther from isostaticity than those from the simulation protocol.

Results for pressure correlations in compressed systems are shown in Fig. 1. In general, we find that $S(q)$ decays isotropically with \mathbf{q} . In Fig. 1(a), we plot the angle-averaged $S(q)$ from simulations, normalized by $K(\Gamma)$ at $\Gamma/A = 10^{-3}$ and 10^{-4} as a function of qD , where D is the small particle diameter. For both frictional $\mu = 1$ and frictionless $\mu = 0$ grains, the results from the simulations match Eq. (4) at small q , with no fitting parameters. In Fig. 1(b) we plot the angle-averaged $S(q)K(\Gamma)$ from experiments. Both simulations and experiments confirm the theoretically predicted scaling of $S(q)$ as $1/K(\Gamma)$ and increasing stiffness as the system is decompressed.

Pure Shear: In the presence of an imposed pure shear, the positivity constraints on the stress lead to different conditions on the stiffness constant K in the x and y directions. A dramatic consequence is that the pressure correlations $S(q)$ become anisotropic even for infinitesimal shear, and the correlations in real space become long-ranged. To lowest order in ψ the entropy functional is

$$\mathcal{L}_{\tau,\Gamma}[\psi] = \int d^2r \left\{ \frac{K(\Gamma + \tau)}{2} (\partial_x^2 \psi)^2 + \frac{K(\Gamma - \tau)}{2} (\partial_y^2 \psi)^2 \right. \\ \left. + \lambda (\partial_x \partial_y \psi)^2 \right\} + K'(\Gamma, \tau) \partial_x^2 \psi \partial_y^2 \psi. \quad (5)$$

Here, x (y) is the principal axis of $\hat{\Sigma}$ with the smaller (larger) eigenvalue. In the case of pure shear, there are now two distinct stiffness coefficients to ensure that both $\hat{\sigma}_{xx}$ and $\hat{\sigma}_{yy}$ are positive. In addition, K' controls the entropy cost of fluctuations that contribute to $\hat{\sigma}_{xx}\hat{\sigma}_{yy}$. A more symmetric version can be constructed by demanding that $K' = \sqrt{(K(\Gamma + \tau)K(\Gamma - \tau))}$, although we have no rigorous argument to support this form.

The pressure correlations predicted from Eq. (5) are:

$$S(\mathbf{q}) = q^4 (K(\Gamma + \tau)q_x^4 + K(\Gamma - \tau)q_y^4 + 2K'(\Gamma)q_x^2 q_y^2) \\ + K(\Gamma)\xi^2 q^6)^{-1}. \quad (6)$$

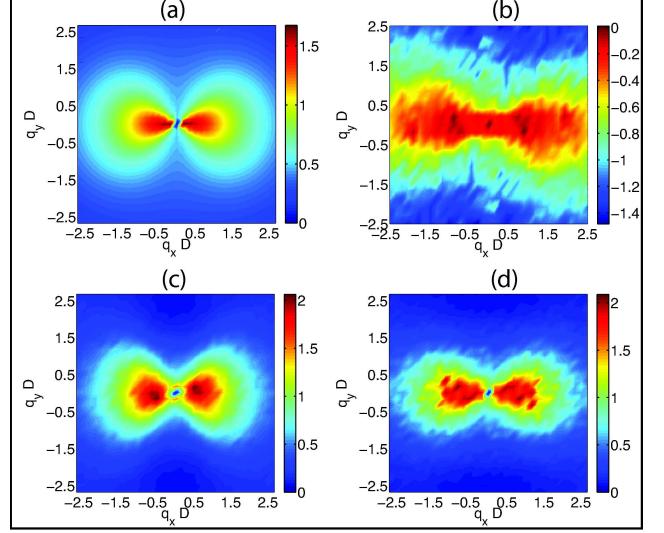


FIG. 2: Contours of $S(q)K(\Gamma)$ under pure shear: (a) theory, using $\tau/\Gamma = \xi/D = 0.3$; (b) experiment, with intensity on a log-scale; and simulations of (c) frictionless and (d) frictional ($\mu = 1$) particles. In both sets of simulations, $\tau/\Gamma = 0.3$. In all plots, compression (dilation) is along the vertical (horizontal) axis.

The anisotropic, dipolar nature of this correlation function is depicted in Fig. 2 (a). To compare theory with experiment, we create a sheared packing by first isotropically compressing the system to a mechanically stable state at a density slightly above jamming. We then apply pure shear by expanding the system in one direction while compressing in the other, keeping the density constant. The resultant pressure correlations are shown in Fig 2 (b), and they match the expected form within the noise of the data. To compare theory and simulation, we generated MS packings of bidisperse disks with and without friction over a range of stress ratios τ/Γ . To do this, we compressed (dilated) the simulation cell in the y (x) direction by $\epsilon = \delta L/L$ over the range $\epsilon = [10^{-5}, 10^{-3}]$. Pressure correlations from simulations in Fig. 2 (c) and (d) also show the dipolar character of the pressure correlations. A key prediction of Eq. 6 is that $\lim_{q \rightarrow 0} S(\mathbf{q})$ depends on the direction of approach. This feature is clearly demonstrated in Fig. 3, which shows the simulation and experimental results for $S(\mathbf{q})$ along different cuts in q -space, along with the small- q predictions from theory. There is good agreement between theory and simulation for $q_x = 0$ and $q_y = 0$, where theoretical predictions exist. Even though the theoretical predictions make several simplifying assumptions such as $z - z_{iso} \ll 1$ (small Γ) and $\tau/\Gamma \ll 1$, we observe qualitative agreement with experimental data. In particular the pressure correlations depend on the direction of approach to $q = 0$ and they are larger along $q_y = 0$ than $q_x = 0$.

The anisotropic nature of correlations in q -space imply anisotropic decays in real space [30] with a slower decay

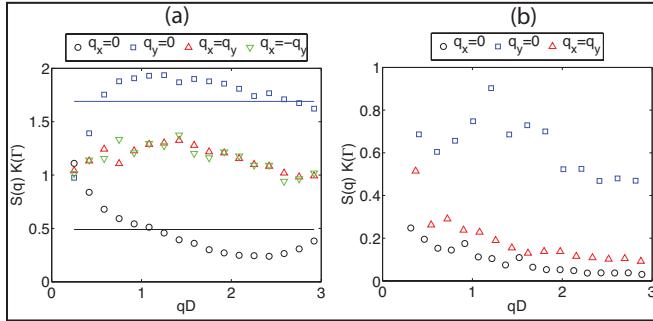


FIG. 3: Cuts along axes specified in the legends for $S(q)K(\Gamma)$ under pure shear. (a) Simulation results with $\mu = 0$ and $\tau/\Gamma = 0.3$. The solid lines are theoretical predictions for $q_x = 0$ and $q_y = 0$. Results for $\mu = 1$ are qualitatively similar. (b) Experimentally measured $S(q)K(\Gamma)$ contours at $\tau/\Gamma = 0.51$. The ratio of $S(q_y = 0)/S(q_x = 0)$ is close to the theoretical prediction in (Eq. 6), $1 + 4\frac{\tau}{\Gamma} + O((\frac{\tau}{\Gamma})^2) \simeq 3$.

along the direction of higher compression. The entropic formulation with the positivity constraint, therefore, provides an explanation for the shear-induced anisotropy in pressure correlations observed in experiments [6].

Discussion: We present a field theoretic approach, based on entropy of packings, for describing stress fluctuations in granular packings. The theory enforces conditions of mechanical stability and positivity of contact forces, and applies close to the jamming transition, where grains have small deformations. From the theory, we calculate pressure correlations and show that they depend sensitively on the method used to generate the jammed state. Under isotropic compression, all correlations are isotropic and obey a simple scaling relation as a function of compression. For packings subjected to pure shear, the correlations are anisotropic with a characteristic dipolar feature in q -space. The anisotropy is a consequence of the positivity constraint, which causes q -space stress fluctuations to be reduced along the compressive direction. The present approach provides a means of relating stress fluctuations to the history of granular systems, which determines the force moment tensor, and an explanation for the anisotropic behavior of stress fluctuations. The theoretical predictions for the pressure correlation functions are confirmed, semiquantitatively, by simulations of MS packings with and without friction and by experiments on photoelastic disks. This agreement is remarkable since it validates the idea that the entropy of MS packings can be used to determine the response of the this far-from-equilibrium system.

Work supported by nsf-dmr0555431 (BC,SH), nsf-dmr0448838 (GL), nsf-dms0835742 (CO), nsf-dmr0555431 (JZ,TSM,RB), and YINQE (GL). BC acknowledges discussions with Nick Read, and CS, BC, GL acknowledge the Aspen Center for Physics and Lorentz Center, where aspects of this work were performed.

-
- [1] C.-h. Liu, S. R. Nagel, D. A. Schecter, S. N. Coppersmith, S. Majumdar, O. Narayan, and T. A. Witten, Science **269**, 513 (1995).
 - [2] J. Zhou, S. Long, Q. Wang and A. D. Dinsmore, Science **312**, 1631 (2006).
 - [3] D. M. Mueth, H. M. Jaeger, and S. R. Nagel, Phys. Rev. E **57**, 3164 (1998).
 - [4] P. Dantu, Ann. Ponts Chaussees **4**, 144 (1967).
 - [5] J. Geng, D. Howell, E. Longhi and R. P. Behringer, G. Reydellet, L. Vanel, E. Clément, and S. Luding, Phys. Rev. Lett. **87**, 035506 (2001).
 - [6] T. S. Majmudar and R. P. Behringer, Nature **435**, 1079 (2005).
 - [7] L. D. Landau and E. M. Lifshitz, Theory of Elasticity, (Butterworth-Heinemann, London, 1986).
 - [8] M. Otto, J.-P. Bouchaud, P. Claudin, and J. E. S. Socolar, Phys. Rev. E **67**, 031302 (2003).
 - [9] M. E. Cates, J. P. Wittmer, J.-P. Bouchaud and P. Claudin, Phys. Rev. Lett. **81**, 1841 (1998).
 - [10] R. Blumenfeld, Phys. Rev. Lett. **93**, 108301 (2004).
 - [11] I. Goldhirsch and C. Goldenberg, Eur. Phys. J. E **9**, 245-251 (2002).
 - [12] A. V. Tkachenko and T. A. Witten, Phys. Rev. E **60**, 687 (1999).
 - [13] C. F. Moukarzel, Phys. Rev. Lett. **81**, 1634 (1998).
 - [14] M. Wyart, S. R. Nagel, and T. A. Witten, Europhys. Lett. **72**, 486 (2005).
 - [15] S. Henkes, C. S. O'Hern and B. Chakraborty, Phys. Rev. Lett. **99**, 038002 (2007).
 - [16] R. Blumenfeld in *Lecture Notes in Complex Systems Vol 8: Granular and Complex Materials*, edited by T. Aste, A. Tordesillas and T. D. Matteo (2007).
 - [17] B. P. Tighe, A. R. T. van Eerd and T. J. H. Vlugt, Phys. Rev. Lett. **100**, 238001 (2008).
 - [18] C. Song, P. Wang and H. A. Makse, Nature **453**, 629 (2008).
 - [19] R. C. Ball and R. Blumenfeld, Phys. Rev. Lett. **88**, 115505 (2002).
 - [20] S. Henkes, Ph.D. dissertation, (2008); S. Henkes and B. Chakraborty, to appear in Phys. Rev. E. (2009).
 - [21] N. Goldenfeld, “Lectures on Phase Transitions and the Renormalization Group”, (Addison-Wesley, New York, 1992).
 - [22] B. Tighe, private communication.
 - [23] Higher-order derivatives of ψ will likely enter if $K \rightarrow \infty$ and suppress stress fluctuations.
 - [24] C. S. O'Hern, L. E. Silbert, A. J. Liu and S. R. Nagel, Phys. Rev. E **68**, 011306 (2003).
 - [25] L. E. Silbert, D. Ertas, G. S. Grest, T. C. Halsey and D. Levine, Phys. Rev. E **65**, 031304 (2002).
 - [26] N. Xu, J. Blawzdziewicz and C. S. O'Hern, Phys. Rev. E **71**, 061306 (2005).
 - [27] H. P. Zhang and H. A. Makse, Phys. Rev. E **72**, 011301 (2005).
 - [28] T. S. Majmudar, M. Sperl, S. Luding and R. P. Behringer, Phys. Rev. Lett. **98**, 058001 (2007).
 - [29] J. Zhang, T. S. Majmudar, A. Tordesillas, and R. P. Behringer, preprint (2009).
 - [30] S. Henkes, unpublished (2009).