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Athermal Fluctuations in Disordered Crystals

Pappu Acharya,[1,*] Surajit Sengupta,[1,†] Bulbul Chakraborty,[2,‡] and Kabir Ramola[1,§]
[1]*Centre for Interdisciplinary Sciences, Tata Institute of Fundamental Research, Hyderabad 500107, India*
[2]*Martin Fisher School of Physics, Brandeis University, Waltham, Massachusetts 02454, USA*

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We analyze the fluctuations in particle positions and interparticle forces in disordered crystals composed of jammed soft particles in the limit of weak disorder. We demonstrate that such athermal systems are fundamentally different from their thermal counterparts, characterized by constrained fluctuations of forces perpendicular to the lattice directions. We develop a disorder perturbation expansion in polydispersity about the crystalline state, which we use to derive exact results to linear order. We show that constrained fluctuations result as a consequence of local force balance conditions, and are characterized by non-Gaussian distributions, which we derive exactly. We analytically predict several properties of such systems, including the scaling of the average coordination with polydispersity and packing fraction, which we verify with numerical simulations using soft disks with one-sided harmonic interactions.

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Introduction.—Disorder in solids can originate from various sources including quenched impurities, polydispersity in particle sizes, as well as their random thermal motion [1–3]. In thermal systems, temperature introduces a natural disorder strength that governs the scale of microscopic fluctuations [4], and consequently controls macroscopic properties. However, many disordered systems when cooled to low temperatures begin to display marked deviations from thermal behavior [5], with temperature playing only a weak role in global properties. Examples of such "athermal" materials include systems displaying glassy behavior [6,7], and jammed packings of particles [8]. Jammed packings arise in a variety of natural contexts and have been the subject of intense scrutiny in recent years [9–12]. At low temperatures such systems are governed purely by the constraints of mechanical equilibrium, with disorder arising from their many possible arrangements. Although their properties have been sought to be modeled within thermal frameworks [13,14], including with temperature-like quantities such as angoricity [15,16], constructing a statistical mechanical theory for such materials has remained elusive. While many studies have focused on the statistical properties of jammed soft particles [17,18], in particular close to the unjamming transition [19–23], a clear understanding of the nature of the jammed phase and its description within a microscopic framework is still lacking. It is therefore important to develop exact theoretical techniques with which to treat such systems.

In this Letter we present exact results for fluctuations and distribution functions in jammed soft particle packings. We show that athermal disorder characterized by polydispersity, induces fundamentally different statistical properties in jammed systems as compared to thermal disorder. In order to make analytic predictions we make use of a well-known paradigm where exact results are obtainable: that of *crystals*. The stability and response of crystals to disorder has been an enduring problem in physics, and several frameworks have focused on thermal fluctuations in crystals, as well as properties of asperities, disinclinations and defects [1,24]. However the properties of crystals composed of jammed particles, where polydispersity introduces an athermal disorder have been relatively less studied [25–27]. We demonstrate that in such athermal crystals the constraints of mechanical equilibrium lead to highly constrained fluctuations of the interparticle forces, in comparison to thermal fluctuations which violate these local constraints. We introduce a disorder perturbation expansion about the crystalline state which allows us to predict several properties of the system including the fluctuations in positions, forces, and bond angles. We use this theory to analytically predict non-Gaussian distributions for the components of forces orthogonal to the original lattice directions, a feature absent from thermal fluctuations.

We consider a system of frictionless disks in two dimensions interacting through a pairwise one-sided potential that is now paradigmatic in the study of soft particles and deformable foams [19,28]. The interaction is given by

$$\begin{aligned} V_{\sigma_{ij}}(\vec{r}_{ij}) &= \frac{\epsilon}{\alpha}\left(1-\frac{|\vec{r}_{ij}|}{\sigma_{ij}}\right)^{\alpha} \qquad \text{for } r_{ij} < \sigma_{ij}, \\ &= 0 \qquad \text{for } r_{ij} \geq \sigma_{ij}. \end{aligned} \tag{1}$$

Here $\vec{r}_{ij} = \vec{r}_i - \vec{r}_j$ is the vector distance between the particles i and j located at positions $\vec{r}_i$ and $\vec{r}_j$, respectively, and $\sigma_{ij} = \sigma_i + \sigma_j$ is the sum of the radii σ_i and σ_j of the two particles. Since the interaction potential only depends on

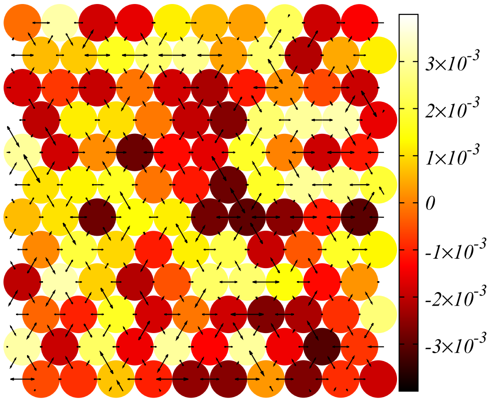

FIG. 1. A section of a disordered crystal composed of jammed soft particles. The particles are colored according to their incremental size $\Delta\sigma_i = \sigma_i - \sigma_0$, where σ_i are their radii and $\sigma_0 = 1/2$. When $\Delta\sigma_i = 0$ the system settles into a triangular lattice. The black arrows represent the change in the interparticle forces from their values in the pure crystal in response to the change in radii. For small disorder (polydispersity) the forces fluctuate primarily along the lattice directions.

the scalar distance $|\vec{r}_{ij}|$, the system can only sustain normal forces. In this Letter, we present results for the harmonic case $\alpha = 2$; however, our techniques can be generalized to systems with different α. The forces are determined by

$$\vec{f}_{ij} = \frac{\epsilon}{\sigma_{ij}}\left(1 - \frac{|\vec{r}_{ij}|}{\sigma_{ij}}\right)^{\alpha-1}\hat{r}_{ij}, \tag{2}$$

where $\hat{r}_{ij}$ is the unit vector along the $\vec{r}_{ij}$ direction. The ground states of the system consist of configurations in mechanical equilibrium; i.e., each particle is in force balance with

$$\sum_j f^x_{ij} = 0, \qquad \sum_j f^y_{ij} = 0, \quad \forall\ i. \tag{3}$$

Here $f^{x(y)}_{ij}$ are the $x(y)$ components of the forces between particles i and j, and the sum extends over all particles j in contact with particle i.

When all the radii are equal, the minimum energy configuration is a crystalline state with the positions of the centers $\{\vec{r}_{i,0}\}$ forming a triangular lattice (see Fig. 1). The distribution of the forces in the crystalline system is given by

$$p(\vec{f}_{ij}) = \frac{1}{6f_0}\delta(|f| - f_0)\delta(\theta - \theta^0_{ij}), \tag{4}$$

where the magnitude of the force f_0 depends on the packing fraction ϕ, and θ^0_{ij} is the angle between the particles i and j in the triangular lattice arrangement. Choosing the equal radii to be $\sigma_i = \sigma_0 = 1/2$, the magnitude f_0 is given by (see the Supplemental Material for details [29])

$$f_0 = 1 - \sqrt{\frac{\phi_c}{\phi}}. \tag{5}$$

Here ϕ_c is the packing fraction of the marginal crystal with no overlaps between particles, with $\phi_c = \pi/\sqrt{12} \approx 0.9069$. The force vanishes for the marginal crystal $\Delta\phi = \phi - \phi_c = 0$.

Thermal versus athermal fluctuations.—We begin by analyzing the differences in the force distributions produced by thermal disorder (characterized by a temperature T), and athermal disorder (characterized by a polydispersity η). For the thermal case, we perform finite temperature Monte Carlo simulations. We begin at the ground state, by creating a triangular lattice of N equal sized disks in a commensurate rectangular box $(L_y = (\sqrt{3}/2)L_x)$. The fluctuations in the positions are then sampled using the interactions given by Eq. (1) at a finite temperature T. As the temperature is increased from $T = 0$ to a finite value, the distribution of the forces deviate from the pure crystalline delta function peaks in Eq. (4), with a mean f_0 and a standard deviation $\propto \sqrt{T}$. This broadening in the force distribution occurs in the components of the forces along the lattice directions $f_{||}$ as well as orthogonal to the lattice directions $f_\perp$. Both these distributions display Gaussian behavior as shown in Figs. 2(a) and 2(b).

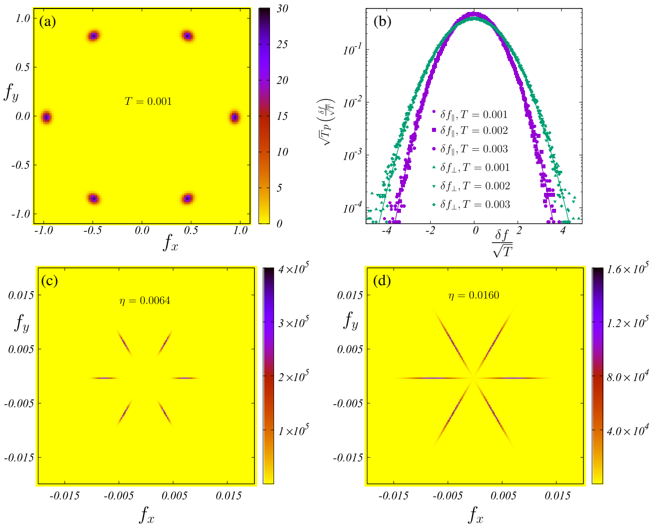

FIG. 2. (a) Two-dimensional distributions of the forces $p(f_x, f_y)$ in the thermal crystal. (b) Scaled distributions of the fluctuations of the components of forces along $(\delta f_{||})$ and orthogonal $(\delta f_\perp)$ to the original lattice directions. Both display Gaussian fluctuations (marked with solid lines) with a variance proportional to the temperature T. (c),(d) In contrast $p(f_x, f_y)$ for the athermal system displays highly constrained fluctuations orthogonal to the original lattice directions as the polydispersity (η) is increased. Here $\phi = 0.92$ and the number of particles is $N = 2500$.

Similarly, we can characterize the fluctuations in forces in the athermal system with increasing polydispersity. In this case the temperature is set at $T = 0$, and the system samples only the ground state for every realization of the disorder, i.e., states in mechanical equilibrium. Disorder is introduced into the system by varying the radii of particles. Starting from the state with all radii equal $\sigma_i = \sigma_0 = \frac{1}{2}$ (i.e., all $\sigma_{ij} = 1$), the radii are incremented as

$$\sigma_i = (1 + \eta\xi_i)\sigma_0, \tag{6}$$

where ξ_i are independent identically distributed (i.i.d.) random variables chosen from an underlying distribution $p(\xi)$. We choose this to be a uniform distribution in the interval $[-\frac{1}{2}, \frac{1}{2}]$ [26]. The polydispersity parameter η quantifies the amount of athermal disorder. For each realization of the noise $\{\xi_i\}$, the system is allowed to settle into a minimum energy configuration as a response to the change in radii. As η is increased from zero, the forces once again deviate from their pure crystalline values. We measure the distribution of the components of forces parallel to the original lattice directions $p(f_{||})$ as the strength of the disorder is increased. This distribution is well fit by a Gaussian with the mean f_0, and standard deviation $\propto \eta$. This seems to suggest that these fluctuations can be modeled by an effective *thermal* Hamiltonian, with the polydispersity playing the role of a temperature $T \propto \eta^2$. However, a striking difference between thermal and athermal fluctuations emerges when one considers the two dimensional distributions of the forces [as shown in Figs. 2(c) and 2(d)]. The distribution of the orthogonal components $p(f_\perp)$ is highly confined with width $\sigma_\perp \ll \sigma_{||}$. On this scale the fluctuations perpendicular to the unperturbed lattice directions are negligible in comparison to the fluctuations along the lattice directions. Since the forces in the system are normal, these constrained fluctuations also imply highly constrained fluctuations in the bond angles θ_{ij}. Moreover, $p(f_\perp)$ displays significant non-Gaussian behavior with increasing polydispersity. Remarkably, as we show below, this distribution can be predicted theoretically. The non-Gaussian nature of this distribution, along with the exact prediction is displayed in Fig. 3.

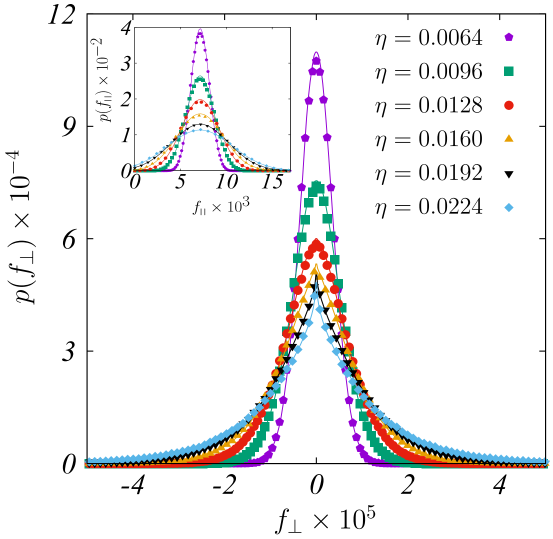

FIG. 3. The distribution of $f_\perp$ in the athermal system, displaying near-perfect agreement with the theoretical prediction in Eq. (12) (solid lines). Here $\phi = 0.92$ and $N = 2500$. This distribution exhibits pronounced non-Gaussian behavior as the disorder strength is increased, in contrast to the thermal system in Fig. 2(b). (Inset) The distribution of $f_{||}$ displays Gaussian behavior. The predictions from the theory are displayed with solid lines.

Disorder perturbation expansion.—In order to theoretically characterize athermal fluctuations, we analyze the response of the crystalline state in the limit of weak disorder. This allows us to treat the polydispersity as a perturbation about the crystalline state. Here we present an outline of the computation, with details provided in the Supplemental Material [29]. The radii in Eq. (6) can be expressed as $\sigma_i = \sigma_0 + \delta\sigma_i$, with $\delta\sigma_i \sim \mathcal{O}(\eta)$. As a response, the positions of the particles deviate from their crystalline values $\{x_{i,0}, y_{i,0}\}$ to a new mechanical equilibrium configuration $\{x_i, y_i\}$. These positions can also be expressed as an expansion in the disorder strength η, which to lowest order is

$$\begin{aligned} x_i &= x_{i,0} + \delta x_i, \\ y_i &= y_{i,0} + \delta y_i. \end{aligned} \tag{7}$$

Here δx_i and δy_i are small perturbations of $\mathcal{O}(\eta)$. The forces in Eq. (2) can then be expressed in terms of these variables as an expansion, which to linear order is given by

$$\begin{aligned} \delta f^x_{ij} &= C^{xx}_{ij}\delta x_{ij} + C^{xy}_{ij}\delta y_{ij} + C^{x\sigma}_{ij}\delta\sigma_{ij}, \\ \delta f^y_{ij} &= C^{yx}_{ij}\delta x_{ij} + C^{yy}_{ij}\delta y_{ij} + C^{y\sigma}_{ij}\delta\sigma_{ij}. \end{aligned} \tag{8}$$

Here $\delta x_{ij} = \delta x_i - \delta x_j$, $\delta y_{ij} = \delta y_i - \delta y_j$ whereas $\delta\sigma_{ij} = \delta\sigma_i + \delta\sigma_j$. The coefficients $C^{\alpha\beta}_{ij}$ can be expressed purely in terms of the positions of the crystalline state, and are translationally invariant. We can exploit this invariance by considering the equations of mechanical equilibrium [Eq. (3)] in Fourier space. Using the forces in Eq. (8), the equations for force balance can be expressed in Fourier space as

$$\begin{pmatrix} A^{xx}(\vec{k}) & A^{xy}(\vec{k}) \\ A^{yx}(\vec{k}) & A^{yy}(\vec{k}) \end{pmatrix} \begin{pmatrix} \delta x(\vec{k}) \\ \delta y(\vec{k}) \end{pmatrix} = \delta\sigma(\vec{k}) \begin{pmatrix} D^x(\vec{k}) \\ D^y(\vec{k}) \end{pmatrix}. \tag{9}$$

Here $\vec{k} \equiv (k_x, k_y) = (2\pi l/2N, 2\pi m/N)$ are the reciprocal lattice vectors of the triangular lattice. The above equation can be interpreted as the change in the position fields in response to the "charges" introduced by the variation in the particle radii. The inversion of this equation in Fourier space yields

$$\delta x(\vec{k}) = \alpha(\vec{k})\delta\sigma(\vec{k}); \quad \delta y(\vec{k}) = \beta(\vec{k})\delta\sigma(\vec{k}). \tag{10}$$

The exact expressions for $\alpha(\vec{k})$ and $\beta(\vec{k})$ are rather cumbersome and we provide a detailed derivation in the Supplemental Material [29].

We are now in a position to derive the fluctuations in the positions of the particles in response to the athermal disorder. For example, the fluctuations in x are given by

$$\langle \delta x_i^2 \rangle = \frac{1}{2L^2} \sum_{\vec{k}} \alpha(\vec{k})\alpha(-\vec{k})\langle \delta\sigma^2 \rangle, \tag{11}$$

where $N = L^2$ is the number of particles in the system. The fluctuations in the radii are i.i.d. variables with $\langle \delta\sigma^2 \rangle = \eta^2/48$. We note that this expression provides the *exact* leading order coefficient of the variance in the positions. In the Supplemental Material [29] we show the excellent agreement between the above theoretical prediction and our numerical simulations.

Non-Gaussian force distributions.—One of the surprising characteristics of athermal fluctuations in disordered crystals is the appearance of non-Gaussian probability distributions in the components of the forces perpendicular to the lattice directions. Remarkably, these distributions can be derived analytically using the perturbation theory in polydispersity as we show below. The fluctuations in the force magnitudes $|f| = \sqrt{f_{||}^2 + f_{\perp}^2} \approx f_{||}$ can be obtained from the position fluctuations using Eq. (8) (see Supplemental Material for details [29]). As the inversion in Fourier space expresses the forces in the system as a linear combination of the fluctuations in the radii, the distribution of $|f|$ can be shown to be a Gaussian with mean f_0 and standard deviation 0.157η. The distribution of $f_{||}$ for various polydispersities is shown in the inset of Fig. 3, along with the theoretically predicted Gaussian distributions showing excellent agreement. Following a similar argument as for $f_{||}$, the fluctuations in the positions can also be used to derive the fluctuations in the bond angles $\delta\theta_{ij} = \theta_{ij} - \theta_{ij}^0$. The distribution of $\sin(\delta\theta)$ is once again a Gaussian distribution with mean 0, and standard deviation 0.0813η. At linear order, the correlations between these variables is small in comparison to their individual fluctuations, and we may treat them as uncorrelated (see the Supplemental Material [29]). These distributions can then be used to derive the distribution of $f_{\perp} = |f| \sin(\delta\theta)$. Since the product of two Gaussian variables with nonzero means exhibits non-Gaussian behavior [34], we find that the distribution of $f_{\perp}$ indeed begins to deviate from a Gaussian distribution at large polydispersities. This distribution is given by (with $s \equiv \sin\delta\theta$)

$$p(f_{\perp}) = \int_0^{\infty} d|f| \int_{-1}^{1} ds\, p(|f|)p(s)\delta(f_{\perp} - |f|s), \tag{12}$$

and can be evaluated analytically [35] (see the Supplemental Material [29]). The pronounced non-Gaussian behavior of the distribution $p(f_{\perp})$ computed using the above expression is displayed in Fig. 3, showing near-perfect agreement with distributions obtained from direct numerical simulations.

Average coordination.—Finally, we use the microscopic predictions from our theory to compute a macroscopic property of the system, namely the dependence of the average coordination on other global parameters such as the polydispersity and packing fraction. Since the magnitude of the forces in the system can only take positive values, the negative regions in the theoretical distribution of $p(|f|)$ represent the broken contacts in the system. Consequently the average coordination to lowest order in η is given by $z = 6\int_0^{\infty} p(|f|)d|f|$. However, we have shown that the distribution of $|f| \approx f_{||}$ is a Gaussian with mean f_0, and standard deviation 0.157η. The dependence of the mean value of the force f_0 on the packing fraction can be obtained by Taylor expanding Eq. (5) up to the first order in $\Delta\phi$, we have $f_0 = \Delta\phi/2\phi_c$. This yields a theoretical prediction for the average coordination in the system

$$z = 3[1 + \mathrm{erf}(\mathcal{C}\Delta\phi/\eta)], \tag{13}$$

with $\mathcal{C}^{-1} = 0.4440\phi_c$. Since all coordination related quantities can be obtained from the underlying force distribution $p(|f|)$, this theory predicts that the average coordination as well as the susceptibilities for different packing fractions can be collapsed with the scaling variable $\eta/\Delta\phi$, as has been observed numerically in previous studies [26]. We plot the variation of the average coordination with polydispersity along with the above theoretical prediction in Fig. 4. Once again this theory does well in tracking the behavior of this nontrivial global parameter, and indeed predicts the scaling with $\eta/\Delta\phi$ perfectly. However, we note that the numerical values of z obtained from simulations display a small deviation from the predicted theoretical curve. We attribute this to the system spanning rearrangements induced by contact breaking events which cannot be exactly modeled within a linear framework. In the Supplemental Material [29] we provide details of this nonlinear contact breaking process observed in the simulations.

Discussion.—In this Letter we have presented exact results for the fluctuations of particle positions and interparticle forces in jammed soft particle crystals. The limit of small disorder allowed us to express the local force balance conditions as a set of linear equations relating the particle coordinates and the particle radii. Exploiting the crystal periodicity of the original lattice, the leading order coefficients of the fluctuations of positions, forces, and relative bond angles could be analytically predicted. This allowed us to express the distribution of the components of the forces perpendicular to the lattice directions as a product of

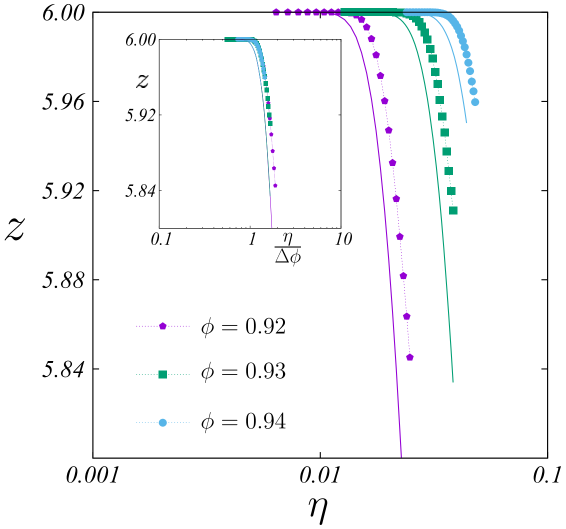

FIG. 4. Variation of the average coordination with the strength of the disorder for different packing fractions (ϕ). The points represent data from simulations, and the solid lines represent the theoretical prediction in Eq. (13). (Inset) The scaling collapse of the average coordination with the predicted scaling variable $\eta/\Delta\phi$. Here $N = 2500$.

two Gaussian variables, which displays non-Gaussian fluctuations.

Since analytic results are rare in the study of disordered jammed matter, it is surprising that many properties of disordered crystals are amenable to theoretical computation. At higher disorder strengths, enough bonds break in the system, and the angular fluctuations become deconfined, which could be considered to be a nonlinear effect. Indeed this system exhibits a nontrivial phase transition to a disordered amorphous phase with increasing disorder [26]. This transition is characterized by diverging fluctuations in coordination numbers over different realizations, and it would be interesting to understand this behavior by studying interactions between defects in the near-crystalline system. Finally, it would also be interesting to use the techniques developed in this paper to predict how microscopic constraints of force balance in such athermal materials give rise to an emergent elasticity at large length scales [36], with nontrivial stress transmission and rheological properties.

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[*] pappuacharya@tifrh.res.in

[†] surajit@tifrh.res.in

[‡] bulbul@brandeis.edu

[§] kramola@tifrh.res.in

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