

Suppressed Compressibility at Large Scale in Jammed Packings of Size-Disperse Spheres

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We analyze the large-scale structure and fluctuations of jammed packings of size-disperse spheres, produced in a granular experiment as well as numerically. While the structure factor of the packings reveals no unusual behavior for small wave vectors, the compressibility displays an anomalous linear dependence at low wave vectors and vanishes when $q \rightarrow 0$. We show that such behavior occurs because jammed packings of size-disperse spheres have no bulk fluctuations of the volume fraction and are thus hyperuniform, a property not observed experimentally before. Our results apply to arbitrary particle size distributions. For continuous distributions, we derive a perturbative expression for the compressibility that is accurate for polydispersity up to about 30%.

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When an assembly of hard particles is compressed, there comes a point where further compression is difficult because the required pressure is too large. A similar “jammed” state can be obtained with soft repulsive particles (as in emulsions or foams), at a particular volume fraction ϕ_c above which the stability of the packing is controlled by the elasticity of the particles. A large body of recent work in both theory and experiment has characterized the properties of jammed packings [1,2].

While much attention has focussed on contacts at the interparticle scale, and force networks and connectivity at larger scales [1–3], there has been comparatively little research into fluctuations and response at very large scales. In Ref. [4] the low-wave-vector behavior of the structure factor, $S(q)$, in a monodisperse system of hard frictionless spheres was studied numerically, revealing “unexpectedly” weak density fluctuations at low q ,

$$S(q) \approx aq, \quad (1)$$

for some constant a , as was also found later for soft particles [5]. This should be compared to the behavior in liquids (including hard sphere fluids at $\phi < \phi_c$), where $S(q) \approx S(q \rightarrow 0) + a'q^2$ [6]. These results imply that bulk fluctuations in the number density are suppressed at ϕ_c : in d dimensions, fluctuations of particle number in subsystems of linear size L scale as $\langle \Delta N^2 \rangle_L \sim L^{d-1}$. Such suppressed density fluctuations are the defining feature of “hyperuniform” materials [7].

Two recent papers reported surprising results, failing to detect the behavior in Eq. (1). A numerical simulation of binary mixtures [8] found that (1) only holds for the particular case of a monodisperse system. Similarly, a confocal microscopy study [9] of a jammed system of moderately polydisperse colloidal hard spheres also failed to observe Eq. (1). Both studies report that $S(q)$ in

size-disperse systems is different from Eq. (1), and suggest that size-disperse packings might not be hyperuniform. That this is a highly topical question is demonstrated by the recent work of Ref. [10], where independently of our approach hyperuniformity was detected, using two-point probability functions.

Here, we report the first experimental observation of vanishing fluctuations of the volume fraction and hyperuniformity in a granular experiment. The same observation is made for numerically produced polydisperse packings with arbitrary size distributions. In contrast with [8,9] we consider not only the structure factor $S(q)$, but also the isothermal compressibility, $\chi_T(q)$, the latter being central to our analysis of size-disperse packings. We propose a novel perturbative approach for extracting $\chi_T(q)$ for continuous size distributions, which in general is a nontrivial task, and explain the connection between vanishing compressibility and hyperuniformity.

First, we briefly describe our systems. Experimentally we produce dense random granular packings by slowly compressing horizontally vibrated bidisperse brass disks. Typically 4500 large disks (diameter 5 ± 0.025 mm) and 3500 small disks (diameter 4 ± 0.025 mm), surrounded by rigid walls, are placed on an oscillating glass plate (amplitude 5 mm, frequency 10 Hz). The packing fraction is increased logarithmically slowly [$d\phi/d \log(t) \approx 10^{-2}$] until the force F measured on the compressing wall increases sharply from $F = 0.05Mg$ to $F > Mg$, where $M = 2$ kg is the typical total mass of the “grains.” At that point the packing jams, grains stop moving and the force measured at the wall remains finite when the vibration is switched off (see Ref. [11] for more details). We take a high resolution picture of the entire packing (2048×2048 pixels) at the largest packing fraction, correcting for optical distortion. We detect the positions of the grains with a resolution of

20 μm and retain ≈ 4000 large grains and ≈ 3000 small grains at least 1 cm away from the walls. We analyze two independent packings, produced starting from uncorrelated initial conditions. Rattler particles are always included in the analysis. Numerically we generate 3D polydisperse sphere packings at ϕ_c using soft repulsive particles, as first proposed in [1], using conjugate gradient methods and small decompression steps to prepare packings exactly at ϕ_c [1,12]. For each set of parameters, we prepare a single, very large configuration composed of $N = 64,000$ particles. We study both a 50:50 binary mixture of spheres with diameter ratio $R \in [1, 2]$, and systems with a continuous size distribution, which we take as a flat distribution centered around the average value $\bar{\sigma}$. We studied polydispersities up to $p = 0.4$, where p is the standard deviation of the size distribution divided by $\bar{\sigma}$.

We now provide some definitions. Consider a size-disperse system composed of n species, containing N_i particles of species i , with diameter σ_i . We define $N = \sum_{i=1}^n N_i$, the density $\rho = N/V$, the concentrations $x_i = N_i/N$, and the partial density fields $\rho_i(\mathbf{q}) = \sum_{j=1}^{N_i} e^{i\mathbf{q}\cdot\mathbf{r}_j}$, where \mathbf{r}_j is the position of particle j belonging to species i . The partial structure factors read $S_{ij}(q) = \frac{1}{N} \times \langle \rho_i(\mathbf{q})\rho_j(-\mathbf{q}) \rangle$, and we collect them in a matrix, $\mathbf{S}(q)$. The total structure factor is defined as usual:

$$S(q) = \sum_{i=1}^n \sum_{j=1}^n S_{ij}(q). \quad (2)$$

When we analyze $S(q)$ in our experimental granular packings, see Fig. 1(a), we find that the low- q behavior is not compatible with Eq. (1). The same observation holds, see Fig. 2(a), for the evolution of $S(q)$ for numerical packings with continuous size distribution of increasing polydispersity. The inset shows that $S(q \rightarrow 0)$ increases continuously with the polydispersity p . This suggests that size dispersity is the main factor responsible for the numerical results in Ref. [8] and the experimental ones in Ref. [9], where $p \approx 0.05$ and $S(0) \approx 0.05$.

For size-disperse systems, we consider not only $S(q)$ but also the compressibility, $\chi_T(q)$. In matrix notation this reads,

$$[\rho k_B T \chi_T(q)]^{-1} = \mathbf{x}^T \mathbf{S}^{-1}(q) \mathbf{x}, \quad (3)$$

where $\mathbf{x}^T = (x_1, \dots, x_n)$. To find χ_T one should measure all partial structure factors in $\mathbf{S}(q)$, and invert this matrix to get $\mathbf{S}^{-1}(q)$. For a one-component (monodisperse) system, one finds $\rho k_B T \chi_T(q) = S(q)$, and both quantities are thus fully equivalent. For a binary mixture, $n = 2$, one gets [13]

$$\rho k_B T \chi_T(q) = \frac{S_{11}(q)S_{22}(q) - S_{12}^2(q)}{x_1^2 S_{22}(q) + x_2^2 S_{11}(q) - 2x_1 x_2 S_{12}(q)}. \quad (4)$$

In Fig. 1(a) we show the compressibility measured experimentally. A clear linear behavior of the compressibility is obtained for low wave vectors. To our knowledge, this anomalous behavior has not been observed experimentally

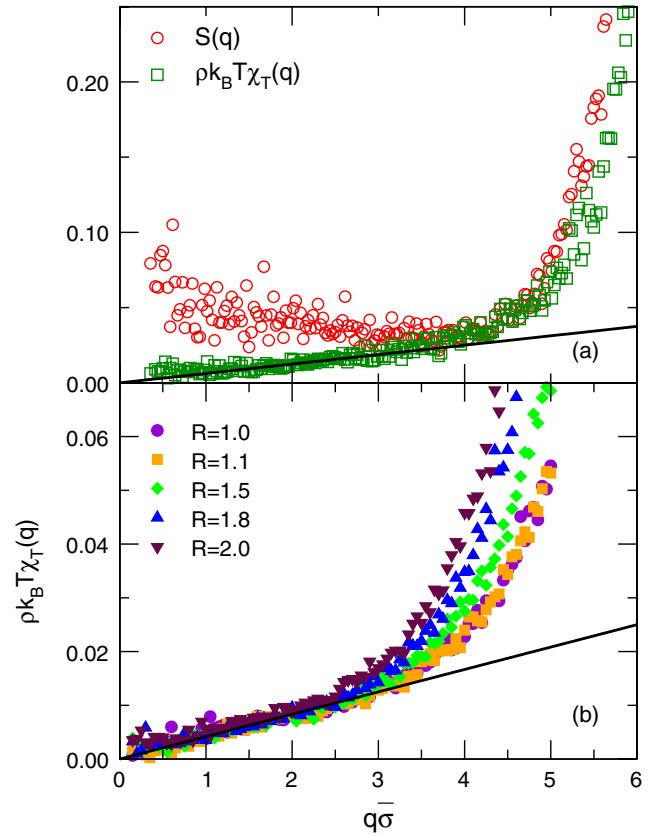


FIG. 1 (color online). (a) Structure factor and compressibility for the two-dimensional packings of disks obtained experimentally. While $S(q)$ resembles that of a binary fluid mixture, the compressibility displays anomalous low- q linear decay. (b) Compressibility for numerically generated jammed packings of 50:50 binary mixtures with various size ratios R . All systems share the same low- q linear behavior of the compressibility.

before. In Fig. 1(b) we show the compressibility obtained numerically, using Eq. (4), for binary mixtures with different size ratio. For all systems considered, a linear behavior of the compressibility is obtained for low wave vectors. This set of results suggests that a relevant generalization of Eq. (1) for binary mixtures is obtained by studying $\chi_T(q)$, rather than $S(q)$.

While straightforward for discrete mixtures with a small number n of components, where \mathbf{S} is an $n \times n$ matrix, the matrix inversion in Eq. (3) is conceptually and computationally difficult for continuous size distributions where the size of the \mathbf{S} matrix formally becomes infinite. The system studied experimentally in [9] is of this type. To analyze such packings, we derive a systematic approximation for $\chi_T(q)$. The idea is that if the size distribution is sufficiently narrow, $\chi_T(q)$ can be obtained perturbatively. To this end, we define $\epsilon_i = (\sigma_i - \bar{\sigma})/\bar{\sigma}$ and derive an expansion of $\chi_T(q)$ in powers of the ϵ_i up to some fixed but otherwise arbitrary order a . To find a suitable starting point for this expansion, recall the relation $S_{ij}(q) = x_i \delta_{ij} + \rho x_i x_j h_{ij}(q)$ between the partial structure factors and $h_{ij}(q)$, the Fourier

transforms of the pair correlation functions $g_{ij}(r) - 1$ [6]. The concentration factors x_i vary rapidly with ϵ_i for narrow distributions, so we cannot expand $S_{ij}(q)$. But the pair correlation functions $g_{ij}(r)$ and hence the $h_{ij}(q)$ depend smoothly on particle size, so we expand the latter up to $\epsilon_i^a \epsilon_j^a$, taking, e.g., for $a=1$, $h_{ij}(q) = h_0(q) + h_1(q) \times (\epsilon_i + \epsilon_j) + h_2(q) \epsilon_i \epsilon_j$ [14]. This expansion is inserted into Eq. (3) for $S_{ij}(q)$, giving after some algebra the compact form

$$[\rho k_B T \chi_T^{(a)}(q)]^{-1} = \mathbf{m}_a^T \mathbf{S}_a^{-1}(q) \mathbf{m}_a, \quad (5)$$

where the matrix $\mathbf{S}_a(q)$ has elements $S^{\mu\nu}(q) = \frac{1}{N} \langle \epsilon^\mu(\mathbf{q}) \epsilon^\nu(-\mathbf{q}) \rangle$, with $\mu, \nu \in \{0, \dots, a\}$, and captures fluctuations of the moment density fields $\epsilon^\mu(\mathbf{q}) = \sum_{i=1}^n \epsilon_i^\mu \rho_i(\mathbf{q})$. Hence, $\epsilon^0(\mathbf{q}) = \rho(\mathbf{q})$ is the number density field, and $S^{00}(q) = S(q)$, the total structure factor. The vector $\mathbf{m}_a^T = (\delta_0, \dots, \delta_a)$ in Eq. (5) has components given by the moments of ϵ_i averaged over the particle size distribution: $\delta_\mu = \sum_{i=1}^n x_i \epsilon_i^\mu$, so that $\delta_0 = 1$, $\delta_1 = 0$, and δ_2 is directly related to the polydispersity, $\delta_2 = p^2$. The result (5) relates the compressibility to the $S^{\mu\nu}(q)$ up to order a . For moderate a it is simple to compute as it only requires the measurement of $(a+1)(a+2)/2$ reduced structure factors. It can be applied to arbitrary particle size distributions and is exact for discrete n -component mixtures if we choose $a = n - 1$, as can be shown by direct calculation from Eq. (3).

We have tested our general formula (5) using computer simulations. When $a = 0$, one has $\rho k_B T \chi^{(0)} = S(q)$, which is only exact for $n = 1$ (monodisperse systems), as discussed above. At first order, $a = 1$, we need to invert a (2×2) matrix to get $\rho k_B T \chi_T^{(1)}(q)$. When applied to the case of a continuous size distribution this formula produces the expected linear behavior at low q for $p \leq 0.10$, but deviations appear at larger p . To check whether these

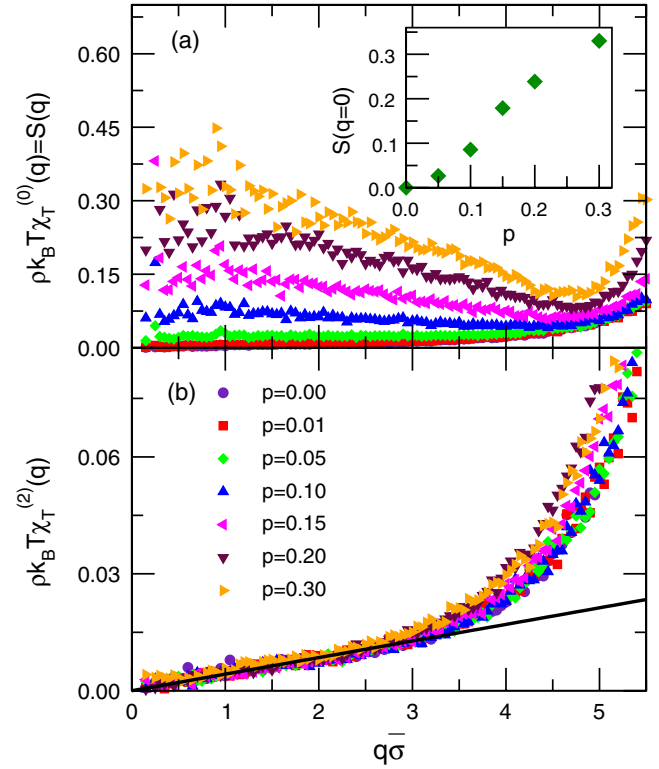


FIG. 2 (color online). (a) Zeroth and (b) second order estimate of the compressibility, Eq. (5), for packings with continuous size distribution of polydispersity p . While the low- q behavior of the structure factor for $a = 0$ is featureless (see inset), the compressibility displays an anomalous linear behavior at low- q , as seen for $a = 2$. The second order estimate of the compressibility can be used up to $p \approx 30\%$, while the first order one (data not shown) becomes unreliable beyond $p \approx 10\%$.

deviations are physical, or a result of our approximation, we go to second order, $a = 2$, where the required inversion of a (3×3) matrix gives:

$$\rho k_B T \chi_T^{(2)}(q) = \frac{S^{00} S^{11} S^{22} + 2 S^{02} S^{01} S^{12} - S^{00} [S^{12}]^2 - [S^{01}]^2 S^{22} - [S^{02}]^2 S^{11}}{S^{11} S^{22} - [S^{12}]^2 + 2 \delta_2 (S^{01} S^{12} - S^{02} S^{11}) + \delta_2^2 (S^{00} S^{11} - [S^{01}]^2)}. \quad (6)$$

We now find, see Fig. 2(b), that a linear q dependence is obtained for polydispersities as large as 30%. This suggests that the same behavior should in fact be obtained for arbitrary size distributions, although measuring the compressibility is more difficult when p is very large because we need to go to even higher orders a .

We now discuss the physical significance of our results. It is perhaps not surprising, with hindsight, that jammed sphere packings have vanishing compressibility since this is precisely how the jamming transition was described in the opening lines of the Letter. However, the quantity we call “compressibility” in this work is in fact a particular combination of density fluctuations, Eq. (3), that only reduces to the compressibility at thermal equilibrium

when the fluctuation-dissipation theorem holds [6]. Remarkably, our results suggest that a similar connection between response and fluctuations may exist far from equilibrium near jamming, even in real granular packings. An obvious connection exists between response and fluctuations at $q = 0$, where $\chi_T(0)$ calculated as a response, $\chi_T(0) = \phi^{-1} \partial \phi / \partial P$, also vanishes on approaching jamming because $P \sim (\phi_c - \phi)^{-1}$. It would be interesting to extend these considerations to finite q near the jamming transition.

Our results also illustrate that, for size-disperse systems, the limit $S(q \rightarrow 0)$ is in general not directly related to the isothermal compressibility, $\chi_T(0)$ [15]. While the latter vanishes in jammed packings, the former is free to take

any positive value. This explains why previous work on size-disperse packings failed to observe any anomalous behavior [8,9]. Both quantities are related to the amplitude of fluctuations of the number density, but $S(q \rightarrow 0) \sim \langle \Delta N^2 \rangle$ captures the total fluctuation of N while $\chi_T(q \rightarrow 0)$ quantifies the fluctuations of N at fixed composition. This can be seen by defining the composition fluctuation fields $c_i(\mathbf{q}) = \rho_i(\mathbf{q}) - x_i \rho(\mathbf{q})$ for $i = 1, \dots, n - 1$. The compressibility from Eq. (3) can then be rewritten as

$$\rho k_B T \chi_T(q) = S(q) - \mathbf{s}_{0c}^T(q) \mathbf{S}_{cc}^{-1}(q) \mathbf{s}_{0c}(q) \quad (7)$$

where the $(n - 1)$ -dimensional vector \mathbf{s}_{0c} gathers the correlations between number and composition fluctuations, and the matrix \mathbf{S}_{cc} the correlations among the latter [13,16]. For the compressibility to vanish at jamming the two terms must cancel, which implies that local fluctuations in N become fully correlated with composition fluctuations. On the other hand, $S(q \rightarrow 0)$ remains positive because a local fluctuation of N can be induced by a local fluctuation of the mixture composition: fluctuations of N do occur in jammed *size-disperse* packings.

The behavior of $S(q)$ in Fig. 2 is clearly inconsistent with Eq. (1), as noticed previously [8,9]. Does this imply that jammed size-disperse packings are not hyperuniform, as suggested in [8]? For point particles, hyperuniformity refers to vanishing bulk fluctuations of the number density, as described in the introduction. However, for an assembly of spherical particles hyperuniformity requires vanishing bulk fluctuations of the local volume fraction [17]. While N and ϕ are directly proportional for monodisperse spheres like those studied in Ref. [4], they are not when the packing is size disperse, and thus no conclusion can be drawn from $S(q)$ alone.

A connection between the anomalous compressibility studied in this work and suppressed fluctuations of the volume fraction in hyperuniform packings can be established. From Eq. (3), we realize that whenever the $\mathbf{S}(q)$ matrix possesses at least one eigenvalue that goes to zero at low q , the compressibility vanishes. We have diagonalized $\mathbf{S}(q)$ or $\mathbf{S}_a(q)$ as obtained in our numerical and experimental packings, and indeed found that in each case anomalous behavior of the compressibility originates from a single vanishing eigenvalue. This implies that there exists a particular linear combination of the partial density fields which has no bulk fluctuations. A detailed analysis of the corresponding eigenvectors shows that they are fully compatible with the local definition of the volume fraction, $\phi(\mathbf{q}) = (\pi/6) \sum_{i=1}^n \sigma_i^d \rho_i(\mathbf{q})$. This identification holds exactly for binary mixtures in our 3D simulations and in the 2D experimental packings. It also holds true for the packings with continuous size distributions, to the same order in ϵ_i that we analyze for the compressibility ($a = 2$, which is accurate up to polydispersity $p = 30\%$). Indeed, we have checked that direct measurements of $I(q) = \langle \phi(\mathbf{q}) \phi(-\mathbf{q}) \rangle$ coincide with the compressibility shown throughout this

article in the low- q regime where linear behavior is observed. Therefore, the anomalous behavior of the compressibility reveals the absence of bulk fluctuations of the volume fraction. This lack of fluctuations was detected also in the recent, independent, study of Ref. [10], using a rather different methodology. We conclude that all our size-disperse jammed packings are hyperuniform.

We have demonstrated anomalous behavior of the compressibility in jammed size-disperse packings of spheres both in simulations and in a granular experiment, using for the case of continuous size distributions an efficient perturbative approach. We have related this to suppressed bulk fluctuations of the volume fraction, or hyperuniformity (see also [10]), thus revealing a structural signature of jamming not seen in the conventional structure factor. Our work also raises intriguing questions about the applicability of fluctuation-dissipation relations to jammed systems.

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