Jamming Transitions in Amorphous Packings of Frictionless Spheres Occur over a Continuous Range of Volume Fractions

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We numerically produce fully amorphous assemblies of frictionless spheres in three dimensions and study the jamming transition these packings undergo at large volume fractions. We specify four protocols yielding a critical value for the jamming volume fraction which is sharply defined in the limit of large system size, but is different for each protocol. Thus, we directly establish the existence of a continuous range of volume fractions where nonequilibrium jamming transitions occur. However, these jamming transitions share the same critical behavior. Our results suggest that, even in the absence of partial crystalline ordering, a unique location of a random close packing does not exist.

Everyday experience shows that it is not possible to compress a disordered assembly of rigid particles beyond a maximal packing fraction. The idea of a critical value for the volume fraction of this "random close packing" has a long history in science [1]. For the idealized case of hard, spherical, frictionless particles the idea recently emerged that a critical value of the volume fraction, named ϕ_J, can be appropriately defined in the thermodynamic limit [2,3], beyond which the system cannot be compressed (alternatively, the system is "jammed") and that it corresponds to a critical point ("point J") with remarkable scaling properties observed when ϕ_J is approached from either side [2,4]. A unique critical packing fraction is also reported under shearing [5,6]. Further studies established the peculiar properties of configurations prepared exactly at ϕ_J, with specific, singular behavior of the pair correlation function [7,8].

The identification of a unique critical point for jamming is surprising because two distinct phenomena occur when compressing hard spheres at equilibrium. First, an equilibrated system of monodisperse hard spheres is fully crystalline above $\phi = 0.54$. Since crystalline states are efficiently packed, it is in principle always possible to increase the volume fraction of a disordered assembly by increasing the local crystalline order, although crystallization is prevented in many studies by using size polydispersity. The protocol dependence of particle arrangements in jammed structures as a relevant consideration has previously been noted and analyzed [9,10].

A second relevant phenomenon is the glass transition. Compressing hard spheres at thermal equilibrium, it is found that ergodicity is lost near a volume fraction much lower than ϕ_J, because the relaxation time for structural relaxation becomes larger than experimental time scales [11]. Necessarily, then, the properties of hard sphere glasses depend on the preparation history, as is well-known for molecular glasses. Specifically, it has been observed in

simulations that liquids in equilibrium sample basins of deeper local energy minima, or inherent structures, at lower temperatures, and the energy of the minima reached at zero temperature upon cooling depends on the cooling rate [12]. Previous numerical work for hard sphere fluids has shown that different compression rates produce glasses with a pressure which appears to diverge at different densities, at least for finite size systems [13–16]. The analogy [17,18] of the jamming density with the energy of inherent structures in systems with soft potentials suggests that the jamming densities should indeed depend both on compression rates as well as on the initial state from which glasses are generated.

The interplay between glass and jamming transitions is currently receiving attention, both from theory and simulations. In mean-field glasses undergoing a random first order transition [19], an exponentially large number of metastable states exist which jam along a continuous, finite, range of densities down to that of the of the "glass close packing" [19,20]. However, none of these mean-field concepts is expected to remain sharply defined in three dimensions [20], and it is important to assess the validity of this interpretation in finite dimensions.

Since evidence is mounting in favor of both a unique jamming transition with critical properties in some papers, or a continuous range of volume fractions in some others, it appears timely to reconcile these two lines of research and answer the following important, open questions. Does a continuous range of volume fractions for jamming necessarily result from crystallization or demixing? Can one reconcile the results found using thermal and athermal protocols? Can the remarkable properties of the jamming transition survive if its location is not unique? Here, we provide precise answers to these questions and close the gap between two sets of ideas.

Recently, a compression protocol was specifically devised to prevent any structural relaxation and crystalline

ordering, thus allowing focusing directly on the sole influence of glassy behavior on jamming [21]. Starting from a well-equilibrated hard sphere fluid configuration at a given volume fraction, ϕ_{init}, one uses a very large compression rate to reach nearly jammed configurations. During these rapid compressions, the glass pressure was observed to diverge within a finite range of volume fractions, even in the thermodynamic limit [21], but the jamming transition was not studied. We shall study numerically how amorphous assemblies of hard spheres prepared using the (thermal) tools of Ref. [21] jam at large volume fraction. We apply the alternative (athermal) tools of Ref. [2] to analyze the properties of jammed configuration at and near the transition. For the specific three-dimensional binary system we use (a 50:50 binary mixture with size ratio 1.4, and periodic boundary conditions), we directly establish the existence of a continuous line of J points extending at least in the range $\phi_J \in [0.648, 0.662]$, all sharing similar critical properties and exponents. Thus, although point J is not unique, its critical properties are.

Particles are first treated as hard spheres and equilibrated using Monte Carlo simulations over a broad range of volume fractions up to $\phi_{\text{init}} = 0.596$, which produces disordered configurations representative of the metastable hard sphere fluid (no crystallization or demixing is observed for simulations as long as 10^{10} time steps). We then use Monte Carlo simulations to rapidly compress these equilibrated fluid configurations. During these compressions the system has no time to relax and retains a structure very close to the initial fluid states [22]. Thus, contrary to previous work employing slow compressions [15,16], we are certain that the jammed configurations we produce contain no more (crystalline) order than the original equilibrated fluid configurations.

When (reduced) pressure is very large, $P/(\rho k_B T) \sim 10^3$, we stop the compression and switch to the athermal procedure of Ref. [2], where particles are now treated as harmonic spheres interacting through a soft pair potential, $V_{\text{soft}}(r_{ij}) = (1 - r_{ij}/\sigma_{ij})^2$. Here, $\sigma_{ij} = (\sigma_i + \sigma_j)/2$ and σ_i is the radius of particle i. The compression then proceeds in a succession of small instantaneous particle inflation followed by energy minimization using conjugate gradient [2]. At large volume fraction, the energy cannot be minimized to zero, and particles overlap: the jamming transition has been crossed. We emphasize that all the details entering this compression protocol in principle quantitatively affect the results presented below [23]. However, since the most sensitive control parameter is ϕ_{init}, we shall only use its value to distinguish between the different protocols, all other parameters being kept fixed. We estimate finite system size effects by studying two system sizes, $N = 1000$ and 8000, chosen in the regime where scaling with N is well understood [2].

In Fig. 1(a) we show the final part (e.g., when particles are treated as soft) of a randomly selected compression history for a system with $N = 10^3$. We follow the energy
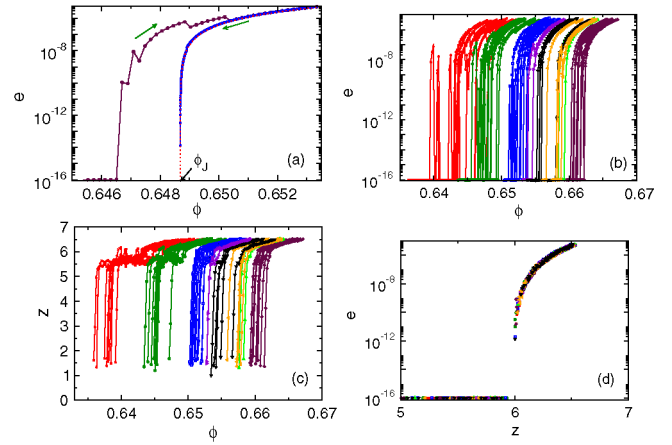

FIG. 1 (color online).   (a) The final part of a compression-decompression cycle across the jamming transition for $N = 10^3$ and $\phi_{\text{init}} = 0.3572$, showing the practical determination of ϕ_J. (b) Evolution of the energy density during compressions of several runs with $N = 1000$. Different curves with the same color correspond to independent realizations at constant ϕ_{init}, while different ϕ_{init} are shown with different colors: $\phi_{\text{init}} = 0.3572$ (red), 0.5397 (dark green), 0.5672 (blue), 0.5744 (purple), 0.5816 (black), 0.5860 (orange), 0.5890 (light green), 0.5935 (maroon). (c) Same as (b) for the evolution of the number of contacts per particle. (d) All data in (b) and (c) collapse when ϕ is eliminated, showing that the jamming transition is better defined by its properties than by its location.

density of the system, $e = N^{-1}\sum_{i<j}V_{\text{soft}}(r_{ij})$, for increasing ϕ. We have defined configurations having energy density less than 10^{-16} as unjammed. For low enough ϕ, we obtain only unjammed particle configurations. Upon increasing the density, there appears a volume fraction above which the energy increases rapidly above zero: this is the jamming transition [2]. Upon further compression, the energy keeps increasing on average, and so does the average number of contacts per particle, z.

This qualitative description becomes more complicated when multiple histories are superimposed [see Fig. 1(b)]. There are several sources of fluctuations in this plot. A first source of fluctuations stems from the possibility for the system to undergo some reorganization which makes the energy drop suddenly, and makes the $e(\phi)$ curve multi-valued during a single compression run. Thus, no simple scaling law for the energy can be detected during compressions. The second source of fluctuations arises when different realizations of the same protocol (i.e., same value of ϕ_{init} but starting from independent fluid configurations) are compared. These fluctuations are present because N is finite, and vanish when $N \to \infty$. A third source of fluctuations can be seen in Fig. 1 when ϕ_{init} is varied, larger ϕ_{init} typically yielding a larger volume fraction for the onset of positive energies.

If we decompress jammed configurations, reorganizations are much less likely, and the $e(\phi)$ curve is smooth enough that it allows a quantitative determination of ϕ_J [see Fig. 1(a)]. Of course, ϕ_J now depends on one addi-

tional parameter, namely, the volume fraction from which decompression starts. We arbitrarily decompress when $e \approx 10^{-7}$ and obtain ϕ_J by fitting e to a power law decay, $e \sim (\phi - \phi_J)^2$ during decompression [2]. Thus, we determine $\phi_J = \phi_J(N, \phi_{init})$. We repeat this analysis for different N, ϕ_{init}, and initial configurations to obtain the statistics of ϕ_J reported in Table I. The main result of this analysis is that $\phi_J(N \rightarrow \infty, \phi_{init})$ seems sharply defined (the fluctuations decrease in good agreement with the expected $N^{-1/2}$ scaling [2]) for each value of ϕ_{init}, but the ϕ_{init} dependence unambiguously survives a substantial increase of system size. Therefore, we have directly established that, for frictionless spheres, the jamming transition occurs along a *continuous line* rather than at a specific point in a volume fraction, as hinted in earlier simulations [16,21], and predicted theoretically [19,20].

Having found several critical points for jamming, rather than one, we now ask whether these distinct J points are equivalent. We have measured the static structure of the jammed configurations through the (partial) pair correlation functions $g_{\alpha\beta}(r)$ (α, $\beta = 1$, 2 for small or large particles, respectively) and the statistics of contacts between particles, $P(z_{\alpha\beta})$. We have also followed the properties of configurations approaching jamming from a large volume fraction. As in [2], we find several scaling laws near our four J points, for P, e, and z. In Fig. 2 we show that for all J points we have $P \sim (\phi - \phi_J)$ and $z = z_c + \sqrt{\phi - \phi_J}$ with $z_c \simeq 6$ (see Table I). Within our numerical precision, therefore, the critical properties of jamming transitions along the J line are identical.

This analysis suggests that the value of ϕ_J is strongly influenced by the entire sample history (compression or decompression protocol, reorganization during compressions, etc.), but that the scaling properties of the jamming transition are much more robust. A simplifying hypothesis is that the history dependence of jammed configurations is mainly contained in the volume fraction ϕ_J at which they jam. This would suggest that data analysis of the jamming

transition should drastically simplify if we eliminate ϕ_J from consideration [24]. We confirm the validity of our hypothesis in Fig. 1(d), where the three sources of fluctuations discussed above are accounted for by eliminating ϕ from the description. By plotting e vs z, we find that data for all cases nicely collapse onto a master curve, independently of the sample preparation history. Thus, the details of the numerical protocols are irrelevant to the relationship between energy and coordination number across the jamming transition.

Not only are scaling properties near jamming robust but the special structure of the pair correlation function at the transition is also observed in all our samples. The pair correlation has a delta peak at contact, which, when integrated, yields the average number of contacts (see Table I). The total number, z, and partial numbers, $z_{\alpha\beta}$, have the same value for all J points, within numerical accuracy. Note, in particular, that z_{12} does not evolve, indicating that demixing between species of the mixture is not observed. In fact the full distributions of contact number barely change [see Fig. 3(a)]. The pair correlation also has a power law singularity near contact [7,8], $g_{\alpha\beta}(r) \sim (r - \sigma_{\alpha\beta})^\gamma$. This is shown in Fig. 3(b) for $g_{11}(r)$, where again the exponent is the same for the four J points, $\gamma \approx -0.5$. In Fig. 3(c), we observe a subtle but systematic evolution with ϕ_{init} of $g(r)$ from near the first minimum to the third peak. Similar to the case of inherent structures in systems with soft potentials [12], these subtle variations reflect the more clearly visible changes in the structure of the equilibrated fluid configurations at ϕ_{init} which were used as starting points for the compressions, as shown in Fig. 3(d). Finally, the increase of ϕ_J does not result simply from a decreasing number of rattlers (particles with no contact), which is instead almost constant (see Table I).

Our results directly establish that a unique location for the jamming transition cannot be expected, even when crystallization or demixing play no role. Rather, our findings are broadly consistent with a "landscape" picture

TABLE I. Statistics of jammed configurations for different system sizes N and various ϕ_{init}. The limit $\phi_J(N \rightarrow \infty, \phi_{init})$ exists, with finite N fluctuations decaying in good agreement with $N^{-1/2}$, and retains a dependence on ϕ_{init}. At the transition, configurations are all isostatic, with number of contacts $z \approx 6$ (with similar distribution width σ_z), and have nearly identical local structure: the number of rattlers, and mean number of contacts between species (z_{12}), are nearly constant.

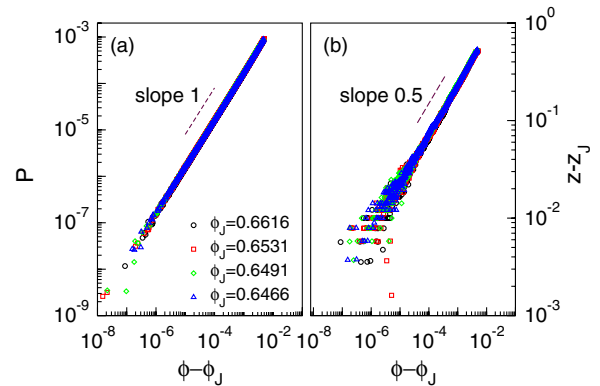| ϕ_{init}	0.3572	0.5397	0.5672	0.5935
ϕ_J (8000, ϕ_{init})	0.6481	0.6499	0.6537	0.6616
ϕ_J (1000, ϕ_{init})	0.6466	0.6491	0.6531	0.6616
σ_J (8000, ϕ_{init})	5.2×10^{-4}	4.9×10^{-4}	3.0×10^{-4}	2.5×10^{-4}
σ_J (1000, ϕ_{init})	9.7×10^{-4}	1.0×10^{-3}	7.6×10^{-4}	5.8×10^{-4}
z at ϕ_J	6.0017	6.0022	6.0021	6.0023
σ_z	2.47	2.50	2.54	2.65
z_{12} at ϕ_J	2.92	2.91	2.92	2.97
Rattlers	4.3%	4.7%	5.4%	6.2%

FIG. 2 (color online). Scaling behavior along the continuous line of J points. The pressure vanishes as (a) $P \sim (\phi - \phi_J)$ while (b) the number of contacts is discontinuous, $z \sim z_c + \sqrt{\phi - \phi_J}$ with $z_c \simeq 6$. We show data for 4 representative points J with 4 different ϕ_J.
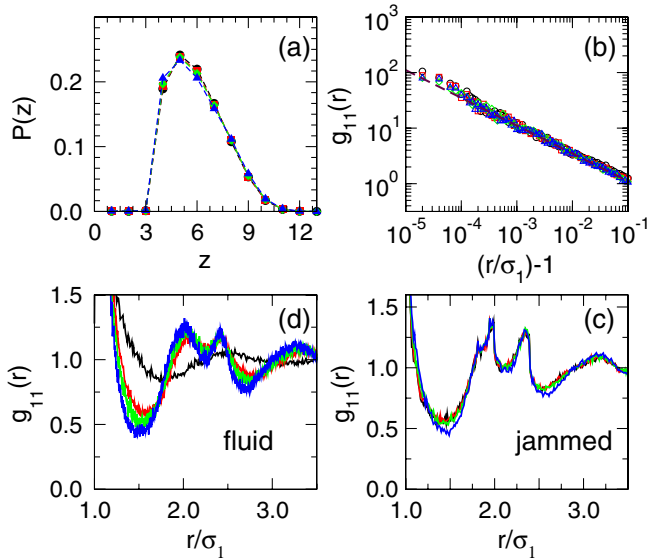
FIG. 3 (color online). (a) Statistical distribution of the number of contacts per particle, z, for 4 points J as in Fig. 2. (b) Power law singularity near contact for $g_{11}(r) \sim (r - \sigma_{11})^\gamma$ for the four points J, with $\gamma = -\frac{1}{2}$ shown as a dashed line. (c) $g_{11}(r)$ near second and third peaks for 4 points J. (d) As in (c) for the equilibrated fluid configurations at ϕ_{init}.

wherein the fluid explores a phase space that does not form the basin of a unique ground state, but instead one marked by the presence of many local minima or metastable states, as is typical of glassy systems. The basins predominantly sampled vary with the equilibrium volume fraction of the system. As in thermal systems [25] using the inherent structure formalism, the nature, number, and evolution with volume fraction of these metastable states can be evaluated [17], and are described exactly in the mean-field limit [19,20]. An evaluation of configurational entropy associated with these jammed states and a comparison with the sharp boundaries predicted in mean-field theory, corresponding to the appearance and vanishing of a configurational entropy, would be valuable for this system.

In summary, we have shown that jamming transitions of amorphous packings of frictionless spheres occur along a continuous range of volume fractions. These transition points are sharply defined in the thermodynamic limit, and this finite range exists even when ordering phenomena or friction are absent, contrasting with the idea of a unique jamming point. Since the location of the jamming transition in fact results from the specific protocol used to study it, we see no reason why it should be reproducible from one experiment to another.

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