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DOI
10.1103/PhysRevLett.121.188002

Publication date
2018

Document Version
Final published version

Published in
Physical Review Letters

Editors' Suggestion

Sticky Matters: Jamming and Rigid Cluster Statistics with Attractive Particle Interactions

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(Received 8 July 2018; published 2 November 2018)

While the large majority of theoretical and numerical studies of the jamming transition consider athermal packings of purely repulsive spheres, real complex fluids and soft solids generically display attraction between particles. By studying the statistics of rigid clusters in simulations of soft particles with an attractive shell, we present evidence for two distinct jamming scenarios. Strongly attractive systems undergo a continuous transition in which rigid clusters grow and ultimately diverge in size at a critical packing fraction. Purely repulsive and weakly attractive systems jam via a first-order transition, with no growing cluster size. We further show that the weakly attractive scenario is a finite size effect, so that for any nonzero attraction strength, a sufficiently large system will fall in the strongly attractive universality class. We therefore expect attractive jamming to be generic in the laboratory and in nature.

DOI: 10.1103/PhysRevLett.121.188002

Numerous complex fluids, including emulsions, foams, pastes, powders, sand, and blood, can jam into soft amorphous solids under increasing packing fraction [1,2]. In recent years, enormous progress towards a fundamental understanding of jammed matter has been driven by theoretical and numerical studies of dense systems of athermal spheres interacting via purely repulsive contact forces. There is now general agreement on how the structure and mechanics of repulsive soft spheres are governed by proximity to the jamming transition at a critical packing fraction ϕ_c—see, e.g., Refs. [3–12] for a partial list. This line of study implicitly (and occasionally explicitly [13]) assumes that repulsive particles yield broad or even universal insights into the marginally jammed state. Nevertheless, purely repulsive interactions are not generic in the laboratory or in nature. While stickiness has various origins (e.g., van der Waals forces [14], depletion effects [15,16], wetting effects [17–19], interface deformation [20,21], critical Casimir forces [22], etc.), particles typically attract their neighbors, and pure repulsion is only possible with careful tuning. The few existing studies of jamming with attraction reveal significant differences, including a gel-like structure with large voids [23,24] and shear banding [25–28]. Most remarkably, Lois *et al.* [29] showed that strongly attractive soft spheres belong to a new universality class, distinct from both repulsive jamming and rigidity percolation on generic lattices [30,31]. But it remains unclear when repulsive jamming gives way to attractive jamming—one cannot currently predict whether a given experimental system falls into the repulsive or attractive jamming class.

In this Letter, we demonstrate that attraction dramatically influences the growth of rigid clusters. A cluster is rigid if, when removed from the packing, its only zero-frequency vibrational modes are rigid body motions. A system is jammed if it contains a spanning rigid cluster [32]. Figure 1 depicts disk packings with "weak" [Fig. 1(a), top row] and "strong" [bottom row] attraction; they differ in the thickness of an attractive shell [Fig. 1(b)]. The largest rigid cluster in each packing is shaded red. For weak attraction, the largest cluster contains just a few particles until a spanning cluster appears suddenly at ϕ_c. This scenario resembles the first-order transition observed in repulsive systems [30,31], suggesting that attraction acts as a small perturbation. In sharp contrast, clusters in strongly

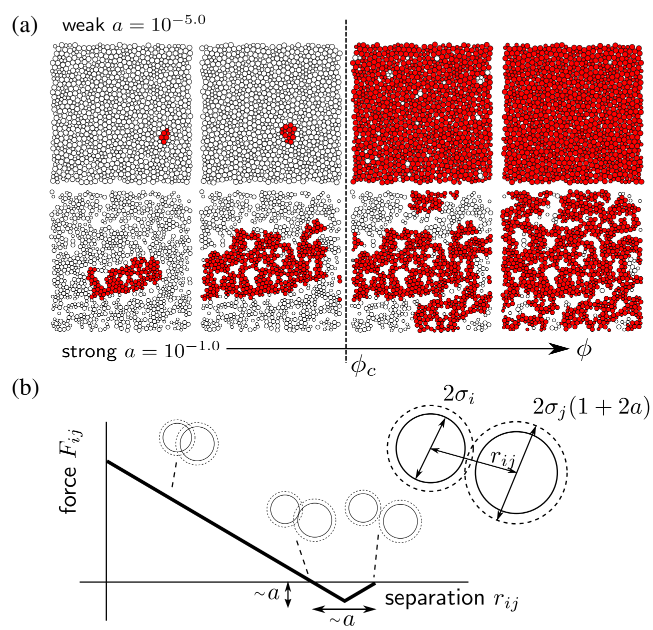

FIG. 1. (a) Packings with weak and strong attraction for packing fractions ϕ near the point ϕ_c where they jam. Particles in red form the largest rigid cluster. (b) Contact force law for a pair of particles with an attractive shell.

attractive systems grow in size before spanning at ϕ_c, reminiscent of a continuous phase transition with a diverging length scale.

What distinguishes repulsive, weakly attractive, and strongly attractive jamming? Here we use rigid cluster decomposition to identify the attractive jamming point and to quantitatively assess the order of the jamming transition. Then, by systematically varying attraction and particle number, we determine when weakly attractive jamming ends and strongly attractive jamming begins. Our central result is that attraction is never weak in the limit of asymptotically large system sizes—large systems are either purely repulsive or strongly attractive, and any amount of attraction places a system in the universality class of strongly attractive jamming.

Methods and protocol.—We consider athermal systems of N disks in a 50:50 bidisperse mixture with size ratio 1.4:1 to avoid crystallization [4,33] and periodic boundary conditions to eliminate wall effects. Unless stated otherwise, we choose $N = 1024$. Because contacts only form and break through external forcing, the structure of both sticky and repulsive packings reflects their history [29,34]. We employ a standard preparation protocol for jammed packings, in which randomly placed particles are instantaneously quenched to a local energy minimum at fixed ϕ using a nonlinear conjugate gradient method [4]. Below, we compare our results to other protocols.

We adopt the conventions of prior work [24–29] and model sticky particles with a repulsive core and attractive shell that experience a central force

$$F_{ij} = \begin{cases} k\delta_{ij} & \delta_{ij} \geq -\sigma_{ij}a \\ -k(\delta_{ij} + 2a\sigma_{ij}) & -\sigma_{ij}a > \delta_{ij} \geq -2\sigma_{ij}a \\ 0 & \delta_{ij} < -2\sigma_{ij}a \end{cases} \quad (1)$$

between particles i and j [see Fig. 1(b)]. The spring constant k characterizes repulsion, while σ_{ij} is the sum of the radii of two cores, r_{ij} is the distance between their centers, and $\delta_{ij} = \sigma_{ij} - r_{ij}$ is their overlap. The dimensionless attraction strength a sets both the attractive shell thickness and the maximal tensile force $-k\sigma_{ij}a$. The packing fraction ϕ is calculated from the particles' cores. Including the attractive shell would increase ϕ by a factor $1 + 4a$, to leading order in a. Note that, unlike repulsive jamming, the jamming point cannot be identified with zero pressure, as tensile states are accessible [23].

The pebble game algorithm [32] efficiently and unambiguously identifies all rigid clusters in two spatial dimensions, dictating our choice to simulate disk packings. The algorithm outputs disjoint sets of bonds (i.e., clusters) whose bonds are rigid with respect to each other. Details are found in Ref. [32]. Accurate contact identification is essential for rigid cluster decomposition. Unlike with repulsive particles, identifying contacts with attraction is straightforward, because particles tend to sit near the first zero of F_{ij} (i.e., the minimum of their pair potential).

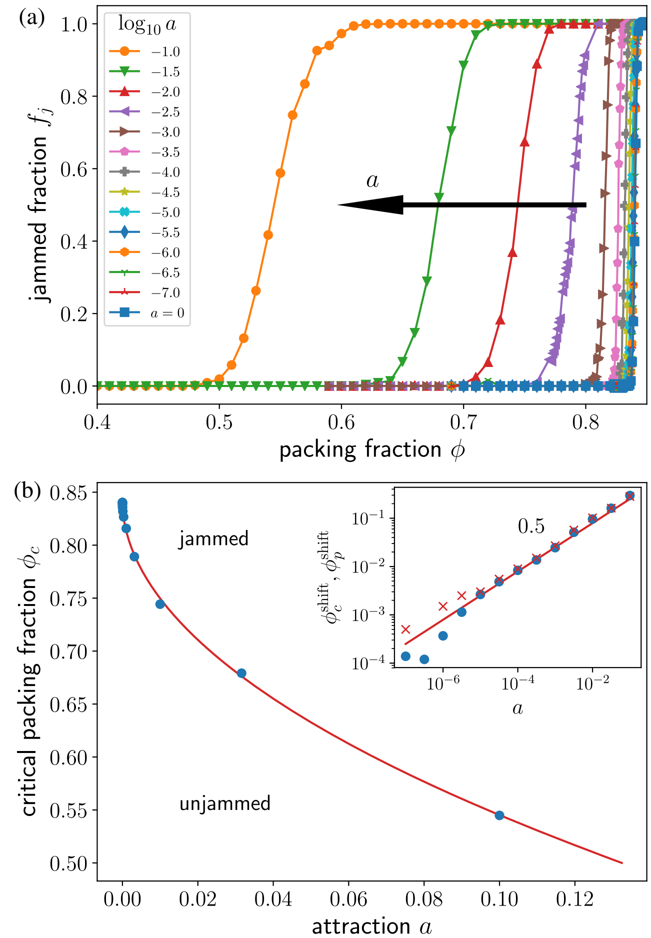

FIG. 2. (a) Fraction of jammed states f_j versus packing fraction ϕ for varying attraction strength a and $N = 1024$. (b) Attractive jamming phase diagram. Inset: Scaling of the shift in $\phi_c(a)$ (filled circles) and $\phi_p(a)$ (crosses) with a.

Jamming phase diagram.—As we are considering physics near jamming, we first determine the critical packing fraction ϕ_c as a function of attraction strength.

For a finite particle number N, the jamming transition is "blurred" by finite size effects, as seen in a plot of the fraction f_j of jammed packings in ensembles prepared at a given ϕ [Fig. 2(a)]. The purely repulsive packings show a rapid increase of f_j at a packing fraction near 0.84. As attraction strength a increases, the rise in f_j shifts to lower ϕ and also becomes more gradual. We will first focus on the shift and then on the widening of f_j.

We associate a critical packing fraction $\phi_c(a, N)$ with the value of ϕ where $f_j(\phi, a, N) = \Delta$, with $\Delta = 0.5$. The shift of the transition is then defined with respect to the purely repulsive jamming point, i.e., $\phi_c^{\rm shift}(a, N) = \phi_c(0, N) - \phi_c(a, N)$. Henceforth, we drop the N dependence of $\phi_c(a)$ whenever $N = 1024$. We have verified that the scaling of $\phi_c^{\rm shift}$ is insensitive to variations in Δ around 0.5.

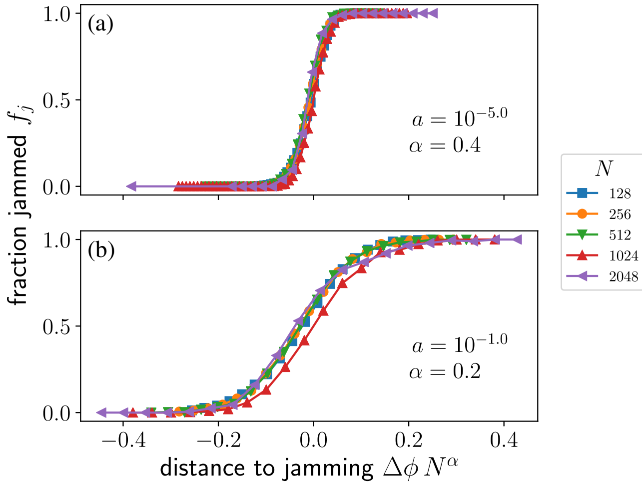

FIG. 3. Data collapse of the fraction of jammed states for varying particle number N in (a) weakly attractive and (b) strongly attractive systems.

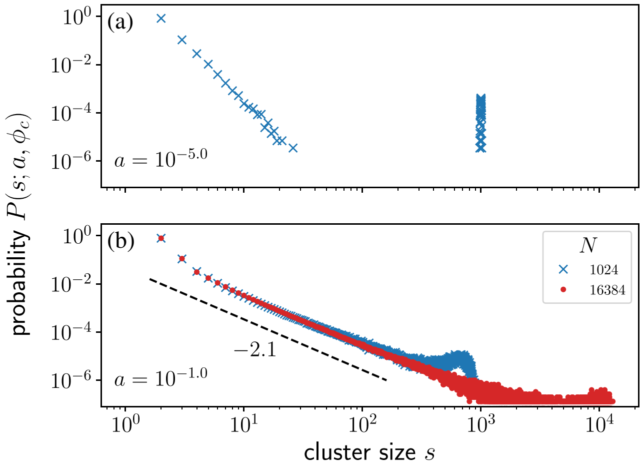

FIG. 4. Cluster size probability distribution for (a) weakly attractive and (b) strongly attractive systems.

In Fig. 2(b), we see how $\phi_c(a)$ decreases with increasing a, dividing the diagram into unjammed and jammed phases. The shift is plotted in the inset of Fig. 2(b) (filled circles). We find power-law scaling that is well described by $\phi_c^{\rm shift} \sim a^{0.5}$. Note that the excess volume occupied by attractive shells, which scales linearly in a, cannot trivially account for this rapid decrease.

We now ask if the jamming transition is sharp in the large system size limit. We focus on "weak" ($a = 10^{-5.0}$) and "strong" ($a = 10^{-1.0}$) attraction, plotted in Figs. 3(a) and 3(b), respectively. For $N = 128...2048$, f_j can be collapsed by plotting versus $\Delta\phi N^{\alpha}$, where $\Delta\phi = \phi - \phi_c(a, N)$. Unscaled data are shown in the Supplemental Material [35]. We observe data collapse for positive values of the exponent α; hence f_j approaches a step function as $N \to \infty$, and the transition is indeed sharp. However, the value of α providing the best collapse for the plotted range of N is different for weak and strong attraction—$\alpha \approx 0.4$ versus 0.2, respectively. This is the first indication in our data of a distinction between weak and strong attraction [36].

Order of the transition.—The growth of rigid clusters seen in Fig. 1 suggests that jamming is a continuous transition in strongly attractive systems, and a first-order transition in weakly attractive and purely repulsive systems. We now quantify these observations by studying the probability $P(s; a, \phi)$ that a given cluster has s particles. From percolation theory, we expect $P(s; a, \phi_c)$ to be gapped in systems with a first-order transition, and to be gapless for a continuous transition [31,37]. Data above and below ϕ_c can be found in the Supplemental Material [35].

The cluster size distribution at ϕ_c is plotted in Figs. 4(a) and 4(b) for weak and strong attraction, respectively. For weak attraction, there is a gap between small clusters of tens of particles or less, and large clusters that contain nearly all particles in the packing, indicating a first-order transition. In the Supplemental Material [35], we verify that the large-cluster peak is solely populated by jammed packings, while small clusters occur in both unjammed and jammed packings.

The cluster size distribution for strongly attractive packings in Fig. 4(b) shows no gap, indicating a continuous transition. We have verified that both jammed and unjammed packings populate the full range of cluster sizes. The distribution has a power-law tail $P \sim s^{-\tau}$ that extends to cluster sizes of order N. To better estimate the exponent τ, we plot the same distribution for a system of $N = 16384$ particles to find $\tau \approx 2.1$ (dashed line). The small peak for s close to $N = 1024$ in the smaller systems is due to finite size effects, including the finite width of f_j. Note that the peak is reduced for larger N, while the distribution remains gapless.

Growing cluster size.—Having addressed statistics at ϕ_c, we now probe cluster size as ϕ is swept through the jamming transition. Our results will further validate the first-order and continuous characterization of weakly and strongly attractive jamming, respectively. Of equal importance, we will also identify the characteristic attraction strength a^* separating weak and strong attraction.

For a continuous percolation transition, one expects to find a typical cluster size that diverges at the transition, while the same quantity should remain finite at a first-order transition [37]. To quantify cluster sizes on either side of jamming, we introduce the probability $n(s; a, \phi)$ that a given nonspanning cluster has s particles and calculate the expected cluster size of a randomly selected particle outside the spanning cluster,

$$\chi(a, \phi) = \frac{\sum_s s^2 n(s; a, \phi)}{\sum_s s n(s; a, \phi)}. \qquad (2)$$

In Fig. 5(a), χ is plotted versus the packing fraction for varying attraction strength. While data for the lowest values

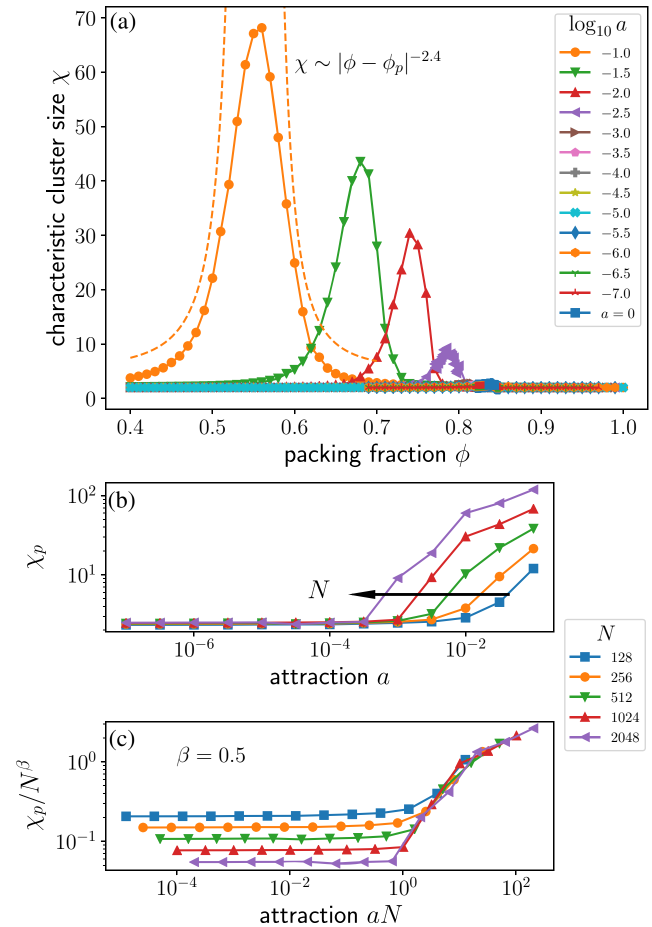

FIG. 5. (a) Cluster size dependence on packing fraction. Dashed line shows the inferred divergence of an infinite system (offset vertically). (b) Evolution of the peak cluster size with attraction strength for varying particle number N. (c) Rescaled data from (b).

of a show no dramatic features, for the strongest attraction strengths there is a substantial increase in χ near ϕ_c. To quantify these observations, we extract the height χ_p and position ϕ_p of the peak in χ. From ϕ_p we calculate the shift $\phi_p^{\text{shift}}(a) = \phi_c(0) - \phi_p(a)$. We find excellent agreement between the position of the peak and ϕ_c determined from Fig. 2(a), as demonstrated in the inset of Fig. 2(b). We conclude that the peak in χ coincides with the jamming point.

We now ask if the peak cluster size diverges as $N \to \infty$. Figure 5(b) shows χ_p as a function of a for varying N. At low a, typical clusters consist of a few particles. There is no trend with N, suggesting that χ_p remains finite. For strong attraction, χ_p grows with N, and the attraction a^* where χ_p starts to grow is lower in larger systems. To gain insight into these effects, in Fig. 5(c) we replot the data as χ_p/N^β versus aN. We observe collapse to a master curve when $aN \gtrsim 1$ and $\beta \approx 0.5$. As β is positive and the master curve increases with aN, we infer that χ_p diverges in the large-system limit—there is indeed a diverging cluster size, consistent with a continuous transition. For the largest a, the cluster size diverges as $\chi \sim 1/|\phi - \phi_p|^{2.4}$ [vertically offset dashed curve in Fig. 5(a); log-log plot in the Supplemental Material [35]].

The rescaled attraction strength aN in Fig. 5(c) indicates that the characteristic scale a^* scales as $a^* \sim 1/N$. Systems with a above (below) a^* jam according to the strongly (weakly) attractive scenario, and any nonzero attraction strength satisfies $a > a^*$ in a sufficiently large system. Our central result follows from this observation: namely, *all* attractive systems jam according to the strongly attractive scenario in the $N \to \infty$ limit, and the purely repulsive limit is singular. In other words, attraction is never a weak perturbation to repulsive jamming.

Discussion.—We have demonstrated that rigid clusters form a jammed phase in purely repulsive and weakly attractive systems via a first-order transition in which the spanning cluster appears suddenly at the critical packing fraction. In sharp contrast, strongly attractive systems jam via a continuous transition with a typical cluster size that diverges at ϕ_c. The first-order transition for weak attraction is a finite size effect, and in thermodynamically large systems the jamming universality class is either purely repulsive ($a = 0$) or attractive ($a > 0$). As attraction is generic in experimental systems, we predict that they jam according to the attractive scenario.

Our packings were prepared using a standard quench protocol [4]. In Ref. [29], Lois *et al.* employed an alternate protocol in which the system is slowly compressed from a dilute state. While they report data for just one attraction strength $a \approx 10^{-2}$, they found a continuous rigidity transition with exponents $\alpha \approx 0.16$ and $\tau \approx 2.1$, in reasonable accord with our $\alpha \approx 0.2$ and $\tau \approx 2.1$. Using yet another protocol, Zheng *et al.* observed a critical packing fraction shift $\phi_c^{\text{shift}} \sim a^{0.3}$, extracted from four values of a over three decades [24]; we find an exponent 0.5 with finer sampling. This reasonable agreement with prior work suggests our results are not specific to packings prepared via a quench. Nevertheless, as with any nonequilibrium transition, an important direction for future work will be to determine how and when protocol dependence occurs. Since sticky contacts tend to persist once formed, we expect protocols can be distinguished by how much they promote contact formation. For example, attractive jammed states can be prepared at an exceedingly low packing fraction by first compressing to a dense state and then decompressing [29].

There are several other likely directions for future work. Foremost, it remains to be determined how rigid clusters influence mechanics, including storage and loss moduli [11,12,38], yield stresses [8,26,27,39–42], nonlocal effects [43–45], and shear bands [26,27,46]. By varying the pair potential, one can also untangle the roles of the range and strength of the attractive interaction. The phase diagram for attractive glasses and gels has ϕ on one axis and the ratio of the attractive well depth U to the thermal scale k_BT on the other. Jammed states at $T = 0$ sit deep in the glass or gel phase; hence one anticipates connections to vitrification or gelation as T increases [47–50].

We acknowledge financial support from the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (Netherlands Organization for Scientific Research, NWO). This work was sponsored by NWO Exacte Wetenschappen (Physical Sciences) through the use of supercomputer facilities.

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[35] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.121.188002 for the following plots: χ_p versus ϕ on a log-log plot, f_j curves without rescaling, and $P(s)$ distributions for varying ϕ on either side of ϕ_c.
[36] Below we present evidence for a crossover $a^* \sim 1/N$ between weak and strong attraction. Hence, for asymptotically large N and nonzero attraction strength, we expect (i) f_j versus $\Delta\phi N^\alpha$ to collapse for the same $\alpha \approx 0.2$, and (ii) the cluster size distribution $P(N_c, \phi_c)$ to be gapless.
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