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Cite this article as:

Xu Ning. Phase Behaviors of Soft-core Particle Systems[J]. *Chinese J. Polym. Sci.*, 2019, 37(11): 1065-1082. doi: 10.1007/s10118-019-2304-2

View online: <https://doi.org/10.1007/s10118-019-2304-2>

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Phase Behaviors of Soft-core Particle Systems

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Abstract This paper reviews some of our recent works on phase behaviors of particulate systems with a soft-core interaction potential. The potential is purely repulsive and bounded, *i.e.*, it is finite even when two particles completely overlap. The one-sided linear spring (harmonic) potential is one of the representatives. This model system has been successively employed to study the jamming transition, *i.e.*, the formation of rigid and disordered packings of hard particles, and establish the jamming physics. This is actually based on the “hard” aspect of the potential, because at low densities and when particle overlap is tiny the potential resembles the hard sphere limit. At high densities, the potential exhibits its “soft” aspect: with the increase of density, there are successive reentrant crystallizations with many types of solid phases. Taking advantage of the dual nature of the potential, we investigate the criticality of the jamming transition from different perspectives, extend the jamming scenario to high densities, reveal the novel density evolution of two-dimensional melting, and find unexpected formation of quasicrystals. It is surprising that such a simple potential can exhibit so rich and unexpected phenomena in phase transitions. The phase behaviors discussed in this paper are also highly regarded in polymer science, which may thus shed light on our understanding of polymeric systems or inspire new ideas in studies of polymers.

Keywords Phase transitions; Soft-core particles; Jamming; Two-dimensional melting; Quasicrystals

Citation: Xu, N. Phase behaviors of soft-core particle systems. *Chinese J. Polym. Sci.* 2019, 37, 1065–1082.

INTRODUCTION

A liquid turns into a solid when being cooled down or compressed. Depending on the rate of cooling or compression, the solid structure will vary. For a sufficiently slow rate, equilibrium phase transition occurs and the solid is crystallized if there are no factors to cause structural frustration. In the other extreme, fast enough rates lead to the glass transition and the liquid turns into a glass with a disordered structure.^[1–3]

It has been well-known that the minimization of free energy is the thermodynamic driving force of crystallization. Normally, the liquid-crystal phase transition is first-order with discontinuous change of volume and entropy. Although it seems true for bulk materials, there are special cases. For instance, due to topological defects, it has been shown that two-dimensional melting or crystallization can be continuous.^[4–6] According to the theory by Kosterlitz, Thouless, Halperin, Nelson, and Young (KTHNY theory), because of the unbinding of dislocation and disclination pairs, there is an intermediate phase named as hexatic phase between liquid and solid with the triangular lattice structure, which makes the transitions from solid to hexatic and from hexatic to liquid to be both continuous.^[7–10] The nature of two-dimensional melting is a hot topic under debate.^[11,12] Some studies have verified the KTHNY theory,^[13–19] while some

others find exceptions.^[4,20–23] Recently, it has been finally settled that for hard disks two-dimensional melting is discontinuous.^[24,25] Moreover, it has also been proposed that the continuity of two-dimensional melting relies on the softness of particles.^[26]

Most of the studies on two-dimensional melting have been focused on crystals with the triangular lattice structure, which has a six-fold rotational symmetry. However, there are other symmetries to form crystals. Apparently, only two-, three-, four-, and six-fold rotational symmetries are allowed. It remains a question whether the two-dimensional melting of crystals with rotational symmetries other than six-fold still exhibit two-stage behaviors.^[27,28] In addition to the issue of two-dimensional melting, one may question whether rotational symmetries other than those listed above are possible in the solid formation. In 1984, Shechtman and coworkers found in the Al-Mn alloy a ten-fold rotational symmetry.^[29] Later, people verified the existence of such solids with crystal forbidden rotational symmetries and named them as quasicrystals.^[30] Quasicrystals have long range order, but do not have periodic structures. Previous studies have shown that quasicrystals are sensitive to conditions. In order to form quasicrystals, one needs to prepare multiple length scales in advance to stabilize the quasicrystal. The length scales reflect in the size,^[31] interaction,^[32–36] or anisotropy of constituent particles.^[37–39] It is always interesting to see quasicrystals in some purposely designed systems. It will be surprising if quasicrystals can emerge in some widely studied systems which do not possess multiple length scales.

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Received April 24, 2019; Accepted May 25, 2019; Published online July 9, 2019

Compared with crystallization, our understanding of the glass transition or the formation of disordered solids is still limited, although the glass transition has been studied and debated for decades. Unlike crystals which have a long range structural order, structures of glasses are disordered and similar to liquids. Until now, there is no widely accepted order parameter to clarify the nature of the glass transition, so it still remains elusive whether the glass transition can be classified into a kind of thermodynamic phase transition or is just a simple kinetic arrest of particles. For more details of the glass transition, there are some good review papers^[1–3] to refer to.

Another widely studied transition closely related to the glass transition is the jamming transition.^[40–47] Strictly speaking, jamming transition refers in particular to the formation of rigid and disordered packings of particles interacting *via* repulsions. The simplest case is the static packing of frictionless spheres, *e.g.*, hard spheres. In the study of jamming, we use the packing fraction $\phi = V_p/V_s$ as the metric of density, where V_p and V_s are volumes (areas in two dimensions) of all particles and the system, respectively. There exists a critical packing fraction ϕ_j . When $\phi < \phi_j$, particles do not interact in the absence of thermal motion, *i.e.*, at zero temperature, so the system is not rigid. When $\phi = \phi_j$, particles are suddenly squeezed by nearest neighbors and the whole packing is jammed. For disordered packings of hard spheres, ϕ_j is the highest packing fraction. For soft particles, the packing fraction can go beyond ϕ_j . When $\phi \geq \phi_j$, the packings are rigid with a percolating force network. Therefore, the sudden formation of rigidity at ϕ_j is named as the jamming transition.

Like the glass transition, the jamming transition is also the transition to disordered solids, but in the absence of thermal energy. It was initially expected that the jamming transition corresponded to the zero-temperature glass transition of purely repulsive particles, so that a generalized jamming phase diagram was proposed to unify various types of transitions from liquids to disordered solids.^[40] In the framework of the jamming phase diagram, the formation of disordered solids or the generally defined jamming can be achieved by lowering the temperature T (*e.g.*, the glass transition), increasing the packing fraction ϕ (*e.g.*, the jamming of granular materials or the glass transition of colloidal systems), or lowering the applied shear stress σ_{xy} (*e.g.*, the halt of shear flows when the shear stress is below the yield stress), as shown in Fig. 1. In the jamming phase diagram, the jamming transition at zero temperature and shear stress (at $\phi = \phi_j$) is denoted as point J . According to the initial jamming phase diagram, point J plays an essential role because both the glass transition and yield stress lines converge to it in the zero temperature and shear stress limits. This is rather inspiring, because then the glass transition might be understood from the nature of the jamming transition at point J , which seems simpler than the glass transition due to the absence of thermal fluctuations. However, things do not turn out the way we want. Although being studied for over two decades, the nature of the “simpler” jamming transition still remains elusive.

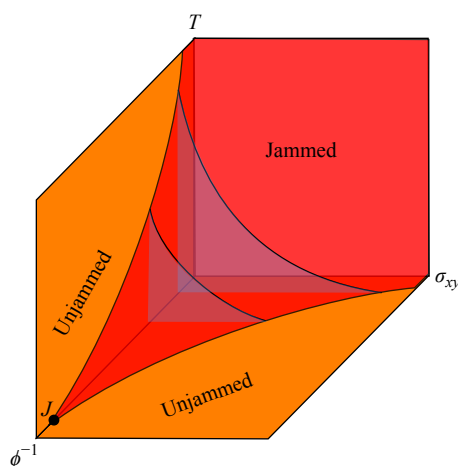


Fig. 1 Initial jamming phase diagram in parametric space of packing fraction ϕ , temperature T , and shear stress σ_{xy} . Both the glass transition and yield stress curves are supposed to converge to point J on the packing fraction axis.

Recently, the initial jamming phase diagram has been further modified. The first modification arose from the doubt whether the jamming transition at point J is indeed the zero-temperature glass transition. It was found that in the zero-temperature limit the glass transition actually occurs at a packing fraction ϕ_g lower than ϕ_j , as shown in Fig. 2 of the modified jamming phase diagram.^[48] The second modification arose from the question whether ϕ_j is a well-defined single value. Recent studies have shown that ϕ_j depends on the preparation history,^[49–53] so that the jamming transition at zero temperature and shear stress is a line instead of a point. Special simulation techniques, *e.g.*, SWAP Monte-Carlo method, can even push the glass transition of hard spheres beyond point J .^[54] Based on the mean field theory, the glass transition of hard spheres can happen over a certain range of packing fractions, which corresponds to the jamming line with point J being the lower packing fraction end.^[49,52,53] These lead to a new phase diagram for hard spheres, as shown in Fig. 3. It has also been proposed that

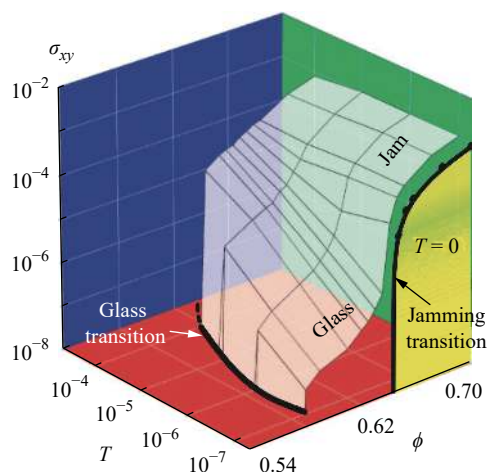


Fig. 2 Modified jamming phase diagram. It shows that the glass transition in the $T \rightarrow 0$ and $\sigma_{xy} \rightarrow 0$ limit occurs at a lower packing fraction than the jamming transition. (Adapted with permission from Ref. [48]; Copyright (2012) American Physical Society)

moving from the glass transition to the jamming transition there is a so-called Gardner transition, which separates stable glasses from marginal glasses. In the perspective of the potential energy landscape, stable glasses are trapped in an energy basin with a single minimum, while for marginal glasses the basin has multiple minima connected by tiny barriers. For more details of recent studies on the Gardner transition, one can refer to the review paper of Ref. [52].

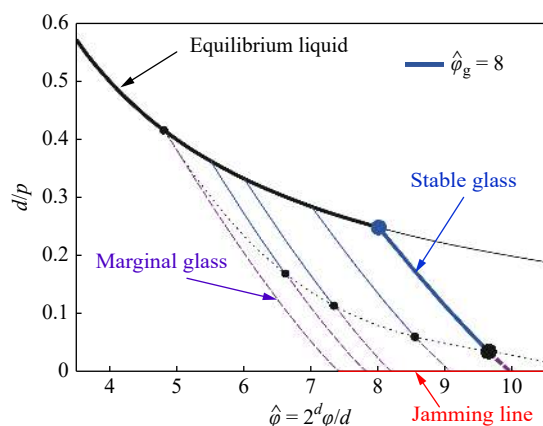


Fig. 3 Modified jamming phase diagram for hard spheres in the inverse pressure d/p and packing fraction $\hat{\phi} = 2^d \phi/d$ plane ($d \rightarrow \infty$), where d is the dimension of space and ϕ in the packing fraction. The black line is the equilibrium liquid line. The black dotted line is the Gardner transition. The red line is the jamming line. (Adapted with permission from Ref. [49]; Copyright (2017) American Physical Society)

Soft matter systems, such as colloidal suspensions and granular materials, have advantages to be model systems to study the fundamental phase transition problems mentioned above and beyond, by treating colloidal particles or grains as atoms. Due to their large sizes (micrometers and above) and slow motion, these particles can be easily tracked. Therefore, colloidal systems are especially ideal for studies of kinetic pathways in phase transitions, which is impossible to study for atomic systems at present. Alternatively, computer simulations are more and more important in studies of phase transitions, relying on the fast growth of the computational power. Being computer “experiments”, simulations have exhibited great power in understanding experimental phenomena, revealing underlying mechanism, verifying theories, and making predictions.

In this feature article, we will summarize our recent progress in the computational studies of phase transitions, which is however not a review of the literature. An interesting aspect is that we employ the same type of particle interaction in all studies. Taking advantage of the special nature of the potential, we are able to attack all issues mentioned above and have interesting and surprising findings. The structure of the paper is as follows. We will first introduce the model and discuss its dual properties. Next, we will focus on low-density systems and show results of the jamming transition and marginally jammed solids. Afterwards, we will show results of high-density systems, including deep jamming, reentrant glass transition, two-dimensional melting, and quasicrystals.

We will conclude in Section V with some perspectives.

MODEL OF SOFT-CORE PARTICLES AND ITS DUALITY

We consider two- and three-dimensional systems with periodic boundary conditions in all directions. The length of the system in each dimension is L . The systems contain N spherical particles with the same mass m . In studies of the jamming and glass transitions, unless specified, we use bidisperse systems composed of $N/2$ large and $N/2$ small particles to avoid crystallization. We set the diameter ratio of the two types of particles to be 1.4. In studies of two-dimensional melting and quasicrystals, all particles are identical.

The particles interact *via* a very simple and purely repulsive potential:

$$U(r_{ij}) = \frac{\epsilon}{\alpha} \left(1 - \frac{r_{ij}}{\sigma_{ij}} \right)^\alpha \Theta \left(1 - \frac{r_{ij}}{\sigma_{ij}} \right) \quad (1)$$

where r_{ij} and σ_{ij} are the separation between particles i and j and sum of their radii, respectively, ϵ is the characteristic energy, α is the parameter to control the particle softness, and $\Theta(x)$ is the Heaviside step function. In all studies, we set the energy scale ϵ , (small) particle diameter σ , and particle mass m to be units of energy, length, and mass. The time is thus in units of $\sigma(\epsilon/m)^{-1/2}$. The temperature is in units of ϵ/k_B with k_B being the Boltzmann constant.

When $\alpha = 2$, Eq. (1) is one-sided harmonic potential, which describes the potential of compressed linear spring. Another widely studied potential is the Hertzian potential with $\alpha = 5/2$, which models typically the interaction between grains for granular materials and is a good approximation to the interaction of poly(*N*-isopropylacrylamide) (PNIPAM) microgels.^[55] Note that the Hertzian potential neglects the deswelling of microgels with the increase of concentration, which can influence phase behaviors.^[56] Of course, the value of α can be freely tuned. In our studies, we concentrate on $\alpha \geq 2$, so that U'' decreases or remains constant (when $\alpha = 2$) with increasing the particle separation. When $\alpha < 2$ and U'' decreases with the decrease of particle separation, some unusual behaviors may happen, *e.g.*, clustering of particles.^[57]

The interparticle potential described by Eq. (1) is soft-core or bounded. When centers of two particles overlap, *i.e.*, $r_{ij} = 0$, the repulsion between the particles is still finite. Therefore, this potential occupies two characters which we can employ: purely repulsive and soft-core. Taking advantage of its repulsive nature, we are able to approach the hard sphere limit. In the low packing fraction regime and in the zero temperature ($T = 0$) limit, particle overlaps are tiny, so the whole particulate system behaves like a hard sphere system. As shown in Fig. 4, harmonic particles have almost identical equation of state to that of hard spheres.^[58] In the high packing fraction regime where particles have to overlap largely, the soft-core nature plays the key role and something interesting happens. For particles with a steep short-range repulsion such as Lennard-Jones ones, particle separations cannot be zero and much more energy is required when two particles are squeezed further. As a result, the melting temperature of the Lennard-Jones solid should always monoton-

ically increase with the density.^[59] However, soft-core particles can completely overlap with finite repulsion, so it is natural to imagine that the melting temperature may have a finite maximum. According to Likos and coworkers,^[60] if the Fourier transform of the soft-core potential is positively definite, reentrant crystallization or melting will occur. As illustrated in Fig. 5, when increasing the density at a fixed temperature, a Hertzian liquid turns into a solid *via* crystallization, but melts again at a higher density.^[28,61,62] This reentrant behavior results in a maximum melting temperature.

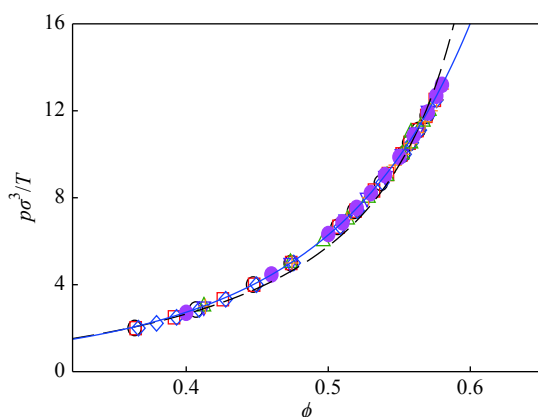


Fig. 4 Equation of state for three-dimensional liquids of hard spheres (solid magenta circles) and soft spheres with harmonic repulsion at several low temperatures. The lines are fittings to the free-volume theory (dashed) and an empirical function (solid). (Adapted with permission from Ref. [58]; Copyright (2009) American Physical Society)

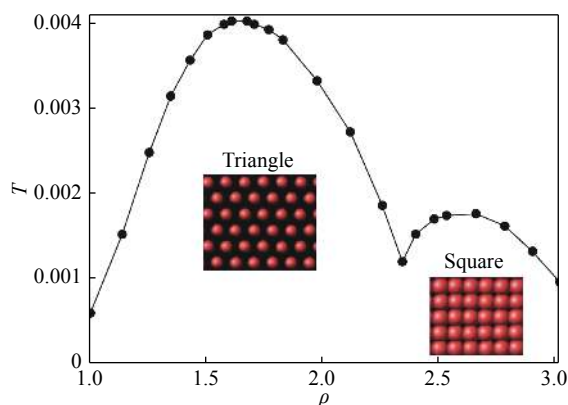


Fig. 5 Phase diagram for two-dimensional Hertzian particles in the temperature T and number density ρ plane. Here, we only show the density region with triangular and square solid structures. There are more structures at higher densities, as shown in Fig. 15. The solid circles are phase boundaries, above which are pure liquid states. The lines are a guide for the eye. The insets are images for triangular and square structures. (Adapted with permission from Ref. [28]; Copyright (2016) American Physical Society)

In the following sections, we will show our studies by focusing on different packing fraction (density) regimes and utilizing the dual nature of the soft-core potential.

We consider both thermal or flowing systems and static packings of particles, *i.e.*, mechanically stable states at $T = 0$. For thermal or flowing systems, we perform molecular dy-

namics simulations.^[63] To get the $T = 0$ states, if not specified, we usually start with random states and quickly find the local energy minima by applying the fast inertial relaxation engine minimization algorithm.^[64] In the presence of shear, we apply Lees-Edwards boundary conditions^[63] instead of periodic boundary conditions to mimic shearing.

JAMMING AT LOW DENSITIES: “HARD” ASPECT OF THE POTENTIAL

In this section, we will concentrate on low packing fraction regimes where particles only slightly overlap and the “hard” aspect of Eq. (1) dominates. We will mainly discuss about our understanding of the criticality of the jamming transition at point J and the generalization of the jamming physics to solids with almost perfect crystalline structures.

Criticality of the Jamming Transition

As a typical transition to disordered solids, the nature of the jamming transition still remains elusive. Original studies have shown special scaling relations in jammed solids of soft spheres interacting *via* the potential of Eq. (1) near point J .^[45]

$$U \sim (\phi - \phi_j)^\alpha \quad (2)$$

$$p \sim (\phi - \phi_j)^{\alpha-1} \quad (3)$$

$$B \sim (\phi - \phi_j)^{\alpha-2} \quad (4)$$

$$G \sim (\phi - \phi_j)^{\alpha-3/2} \quad (5)$$

$$z - z_{\text{iso}} \sim (\phi - \phi_j)^{1/2} \quad (6)$$

where U , p , B , G , and z are the potential energy, pressure, bulk modulus, shear modulus, and average coordination number per particle, and $z_{\text{iso}} = 2d$ is the isostatic value with d being the dimension of space. These relations hold not only for point J , but also for any points on the jamming line as the onset of jamming transition.^[51] Except for z , scaling relations of other quantities rely on potential (α). Moreover, these scalings exhibit both continuous and discontinuous features. For example, z jumps discontinuously from 0 to z_{iso} at point J , while other quantities grow continuously, except that B jumps discontinuously from 0 to a constant value when $\alpha = 2$.

The most special feature of the jamming transition is the isostaticity.^[65] Right at the jamming transition, $z = z_{\text{iso}}$. For a system of N particles in d dimensions, the numbers of constraints and degrees of freedom are $Nz/2$ and dN , respectively. According to the Maxwell criterion, in order to maintain stability, $Nz/2 \geq dN$, leading to $z \geq 2d = z_{\text{iso}}$. Therefore, isostaticity is the minimum requirement of mechanical stability, and jammed solids at point J are on the verge of instability. Disordered solids right above point J are thus termed as marginally jammed solids, because of their marginally stable nature.^[41,42,66]

The scalings shown above imply the criticality of point J , but the criticality, if any, may be different from the well-known ones. There have been quite a few studies aiming at figuring out such a criticality,^[45,67–78] but no quantitative

agreement has been achieved yet. For hard particles, the criticality can only be probed below the jamming transition.^[72,73] By contrast, soft particles gain the freedom on either side of point J .^[68,70] In the following, we will discuss about our approaches in both T - ϕ and ϕ - σ_{xy} planes of the jamming phase diagram, and by using self-propelled soft particles as well.

Jamming-like transition

As mentioned above, the jamming transition at point J is not the $T = 0$ glass transition. Then, in the T - ϕ plane, is there any transition at $T > 0$ converging to point J in the $T = 0$ limit and hence signifying the criticality? One difficulty in understanding the transition from liquids to disordered solids is the lack of structural signature. Earlier studies reveal that there is actually a signature of the jamming transition in the pair distribution function $g(r) = \frac{L^d}{(N/2)^2} \langle \sum_i \sum_{j \neq i} \delta(r - r_{ij}) \rangle$, where d is the dimension of space, the sums are over all large particles for bidisperse systems concerned here, and $\langle \cdot \rangle$ denotes the ensemble or time average. On approaching point J from either side, the height of the first peak of $g(r)$ for static packings, denoted as g_1 , diverges.^[79]

Later, we find that in thermal systems, such as suspensions of PNIPAM microgels, g_1 also reaches a maximum $g_1^{\max}$ at a certain packing fraction $\phi_{j,T}$ or pressure $p_{j,T}$, when the temperature is fixed.^[55,69] With the increase of temperature, $g_1^{\max}$ decreases according to the scaling $g_1^{\max} \sim T^{-0.9/\alpha}$, while $\phi_{j,T}$ moves away from ϕ_j following the relation:

$$\phi_{j,T} - \phi_j \sim (p_{j,T}/\phi_j^2)^{1/(\alpha-1)} \sim T^{1/\alpha} \quad (7)$$

Interestingly, $p_{j,T}$ separates the isothermal curves of concerned quantities into two regimes. When $p > p_{j,T}$, some $T = 0$ jamming scalings, e.g., Eqs. (2) and (3), maintain, while they break down when $p < p_{j,T}$. Note that systems on both sides of $p_{j,T}$ are disordered solids (glasses). The pressure $p_{j,T}$ acts as if a transition point from a type of glass to another type with the jamming features. We thus name the transition at $p_{j,T}$ as the jamming-like transition.^[69] We will come back later to address the underlying difference between the two types of glasses on the two sides of $p_{j,T}$.

The jamming-like transition converges to point J in the $T = 0$ limit. One significance of the transition can be seen from Fig. 6. By combining the scaling of Eq. (7) and the jamming scalings, we come up with the following scaling function:

$$\Gamma = T^{v_\Gamma} f_\Gamma \left(\frac{p}{\phi^2 T^{\frac{\alpha-1}{\alpha}}} \right) \quad (8)$$

Here, Γ denotes concerned quantities such as g_1 , $\phi - \phi_j$, U , and the fluctuation of the coordination number $\delta z^2 = \langle (z - \langle z \rangle)^2 \rangle$ with $\langle \cdot \rangle$ being the time average. The exponent v_Γ can be easily determined for each quantity. Interestingly, Fig. 6 shows that all quantities calculated at different temperatures collapse nicely onto the same master curves. Eq. (8) is a typical mathematical presentation of the scaling hypothesis of critical phenomena.^[80] The scaling collapse following it implies that the state point at $T = 0$ and $p = 0$ is critical. For this case, it is just point J . Therefore, our finding

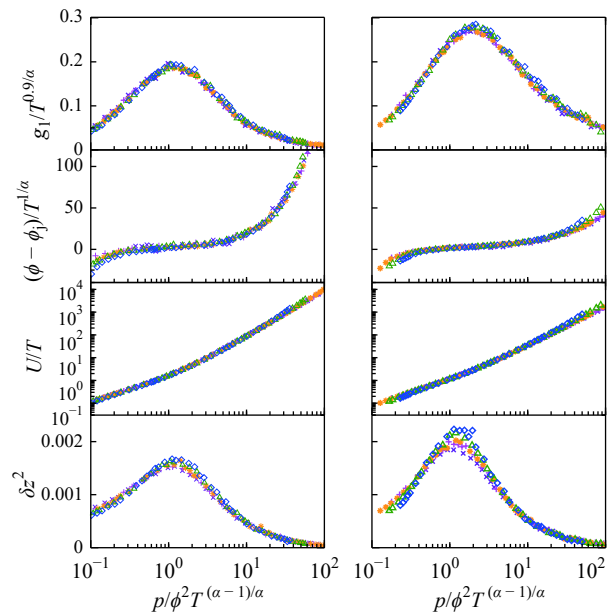


Fig. 6 Scaling collapse of the height of the first peak of $g(r)$, g_1 , packing fraction distance from the $T = 0$ jamming transition, $\phi - \phi_j$, potential energy, U , and fluctuation of coordination number, δz^2 , as a function of pressure p for several low temperatures. Left and right columns are for harmonic ($\alpha = 2$) and Hertzian ($\alpha = 2.5$) systems, respectively. (Adapted with permission from Ref. [69]; Copyright (2013) The Royal Society of Chemistry)

and characterization of the jamming-like transition support the criticality of point J .

With the finding of the jamming-like transition, we propose a new jamming phase diagram in the T - p plane,^[81] as shown in Fig. 7. As mentioned above, the glass transition does not converge to point J in the $T = 0$ limit, unlike what is expected by the initial jamming phase diagram. Now we see that there is actually a transition other than the glass transition converging to point J . Therefore, the jamming-like transition may reminisce some special properties of point J and is thus a possible demarcation of two distinct glasses.

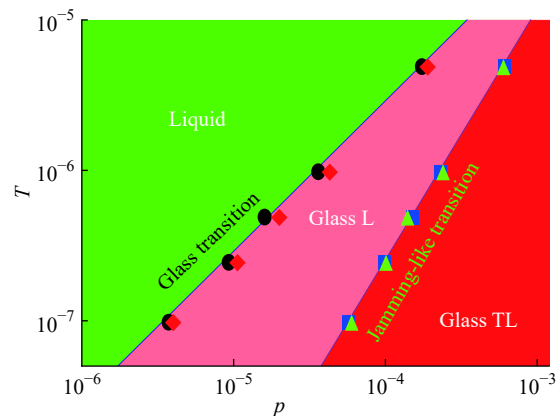


Fig. 7 Phase diagram including the glass transition (circles) and the jamming-like transition (squares) for two-dimensional Hertzian systems. The diamonds and the triangles locate the crossover temperatures where the longitudinal and transverse Ioffe-Regel frequencies drop to zero, respectively. (Adapted with permission from Ref. [81]; Copyright (2015) American Physical Society)

The normal modes of vibration, *i.e.*, phonons, are fundamental to understanding the properties of normal solids like crystals. As the disordered counterpart of crystals, glasses exhibit distinct vibrational properties, which result in unusual thermodynamic and energy transport behaviors.^[41,47,66,78,82–85] It is well known that the structural disorder leads to the localization of the modes.^[82,86–88] Disorder is everywhere in glasses, which thus affects modes at almost all frequencies and more or less destroy the phononic nature of the modes. Phonons are quasi-particles describing the elementary excitation of all constituent particles in crystals. According to the Ioffe-Regel criterion,^[89] phonons are well defined as “particles” if their mean free path exceeds their half wavelength. Due to the presence of disorder, some vibrational modes are strongly scattered and thus lose their particulate nature. There exists an Ioffe-Regel frequency ω_{IR} above which vibrational modes cannot be defined as phonons.

The scattering of the excitations can be clearly seen from the dynamic structure factor^[90,91]

$$S_{\lambda}(k, \omega) = \frac{k^2}{2\pi N \omega^2} \int_0^{\infty} dt \langle \vec{J}_{\lambda}(k, t) \cdot \vec{J}_{\lambda}(-k, 0) \rangle \exp(i\omega t) \quad (9)$$

where λ denotes *T* (transverse) or *L* (longitudinal), ω is the frequency of the excitation, and

$$\vec{J}_{\text{L}}(k, t) = \sum_j [(\vec{v}_j(t) \cdot \hat{k}) \hat{k}] \exp[i\vec{k} \cdot \vec{r}_j(t)] \quad (10)$$

$$\vec{J}_{\text{T}}(k, t) = \sum_j [\vec{v}_j(t) - (\vec{v}_j(t) \cdot \hat{k}) \hat{k}] \exp[i\vec{k} \cdot \vec{r}_j(t)] \quad (11)$$

where the sums are over all particles, $\vec{v}_j$ is the velocity of particle j , and $\vec{k} = k\hat{k}$ is the wave vector. For perfect crystals at $T = 0$, $S_{\lambda}(k, \omega) \neq 0$ occurs only at a certain $\omega = \Omega_{\lambda}$ when k is given. The one-to-one correspondence between Ω_{λ} and k determines the dispersion relation $\Omega_{\lambda} = c_{\lambda}k$ with c_{λ} being the speed of sound. In the presence of disorder, S_{λ} is broadened, as shown in Figs. 8(a) and 8(b). However, $S_{\lambda}(\omega)$ still shows a maximum at a given k , which can be nicely fitted with^[91]

$$S_{\lambda}(k, \omega) = S_{\lambda, \text{B}}(k, \omega) + S_{\lambda, \text{R}}(k, \omega) \\ = \frac{A_{\lambda}(k)}{[\omega^2 - \Omega_{\lambda}^2(k)]^2 + \omega^2 \Gamma_{\lambda}^2(k)} + \frac{B_{\lambda}(k)}{\omega^2 + (D_{\lambda} k^2)^2} \quad (12)$$

where $S_{\lambda, \text{B}}(k, \omega)$ and $S_{\lambda, \text{R}}(k, \omega)$ are the Brillouin and Rayleigh components, $A_{\lambda}(k)$ and $B_{\lambda}(k)$ are fitting parameters, $\Gamma_{\lambda}(k)$ characterizes the broadening of the excitations, and D_{λ} is the thermal diffusivity. Figs. 8(c) and 8(d) show $\Omega_{\lambda} \sim k$ and $\Gamma_{\lambda} \sim k^2$. Ioffe-Regel limit is reached when $\Omega_{\lambda}(k_{\text{IR}}^{\lambda}) = \pi \Gamma_{\lambda}(k_{\text{IR}}^{\lambda})$, from which we obtain the transverse and longitudinal Ioffe-Regel frequencies $\omega_{\text{IR}}^{\text{T}} = \Omega_{\text{T}}(k_{\text{IR}}^{\text{T}})$ and $\omega_{\text{IR}}^{\text{L}} =$

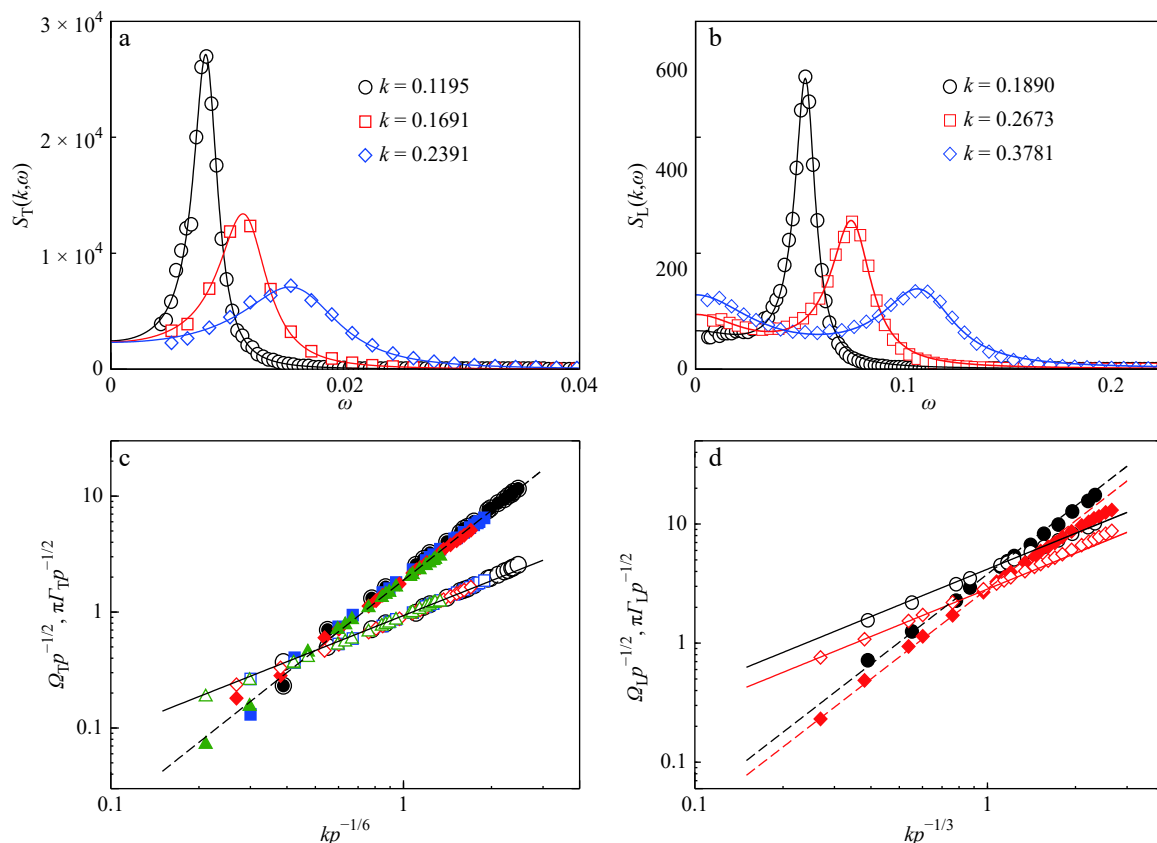


Fig. 8 (a, b) Examples of the transverse and longitudinal dynamical structure factors, $S_{\text{T}}(k, \omega)$ and $S_{\text{L}}(k, \omega)$, for two-dimensional marginally jammed solids with Hertzian repulsion at a low pressure. The lines are fits to Eq. (12). (c, d) Pressure scaled dispersion relation $\Omega_{\text{T(L)}}(k)$ (empty symbols) and broadening of the excitations $\pi \Gamma_{\text{T(L)}}(k)$ (solid symbols) calculated for different pressures. The solid and dashed lines show $\Omega_{\text{T(L)}} \sim k$ and $\pi \Gamma_{\text{T(L)}} \sim k^2$, respectively. (Adapted with permission from Ref. [81]; Copyright (2015) American Physical Society)

$\Omega_L(k_{\text{IR}}^L)$. We find that ω_{IR}^T and k_{IR}^T tend to vanish at the jamming-like transition, while ω_{IR}^L and k_{IR}^L vanish at the glass transition (see Fig. 7).

As shown in Fig. 7, the jamming-like transition separates two types of glasses. On the higher pressure side of the jamming-like transition, the glasses can carry both transverse and longitudinal phonons, so we denoted them as Glass TL. On the lower pressure side, the glasses can only carry well-defined longitudinal phonons because $\omega_{\text{IR}}^T = 0$, so we call them Glass L. In the $T = 0$ limit, Glass L is just the hard sphere glass. Our results imply that hard sphere glasses below the jamming transition do not contain well-defined transverse phonons.

Finite size scaling

True phase transitions occur only in the thermodynamic limit, *i.e.*, when the systems are infinitely large.^[80] In finite size systems, the transition point shifts with system size. Critical phenomena can then be characterized by the finite size scaling. In this section, we will show our finite size analysis of the criticality of point J . Here, we mainly focus on the ϕ - σ_{xy} plane at $T = 0$ in Fig. 1.

Disordered solids can sustain external loads such as shear or compression. When the applied shear stress is smaller than the yield stress σ_y , the system remains solid-like. Otherwise, it will evolve into a shear flow. The boundary of the proposed jammed regime in the ϕ - σ_{xy} plane of the jamming phase diagram is thus the yield stress. In previous approaches, yield stress is estimated by increasing the shear strain γ quasistatically until the system yields. Therefore, the shear

strain is well controlled, but not the shear stress. Note that the shear stress and shear strain are conjugate variables. We are thus able to have the shear stress under control by applying the Legendre transformation to the potential energy U and construct a thermodynamic-like quantity $H = U - \sigma_{xy}\gamma L^d$ with d being the dimension of space.^[71]

At fixed packing fraction ϕ and applied shear stress σ_{xy} , we minimize H for a number of ideal gas states and find the probability $P(\phi, \sigma_{xy})$ of finding jammed states, from which the yield stress σ_y is determined. As shown in Fig. 9(a), for three-dimensional systems of N particles interacting *via* harmonic repulsions, there is a scaling behavior of the yield stress

$$\sigma_y(\phi, N) = \sigma_{0,N}(\phi - \phi_{j,N}) \quad (13)$$

well above the jamming transition, where

$$\phi_{j,N} = \phi_{j,\infty} - 0.137N^{-0.457} \quad (14)$$

$$\sigma_{0,N} = \sigma_{0,\infty} + 0.082N^{-0.363} \quad (15)$$

with $\phi_{j,\infty} = 0.649$ and $\sigma_{0,\infty} = 0.018$ being the critical packing fraction of the jamming transition and yield stress amplitude in the thermodynamic limit. Interestingly, $\sigma_y(\phi, N)$ exhibits the finite size scaling

$$\sigma_{yn} = \frac{\sigma_y}{\sigma_{0,N}} = (\phi - \phi_{j,\infty})g_\sigma[(\phi - \phi_{j,\infty})N^\mu] \quad (16)$$

with a limiting behavior $\sigma_y \sim (\phi - \phi_{j,\infty})$, where $\mu = 0.41 \pm 0.02$, as shown in Fig. 9(b). This finite size scaling implies the existence of a length scale

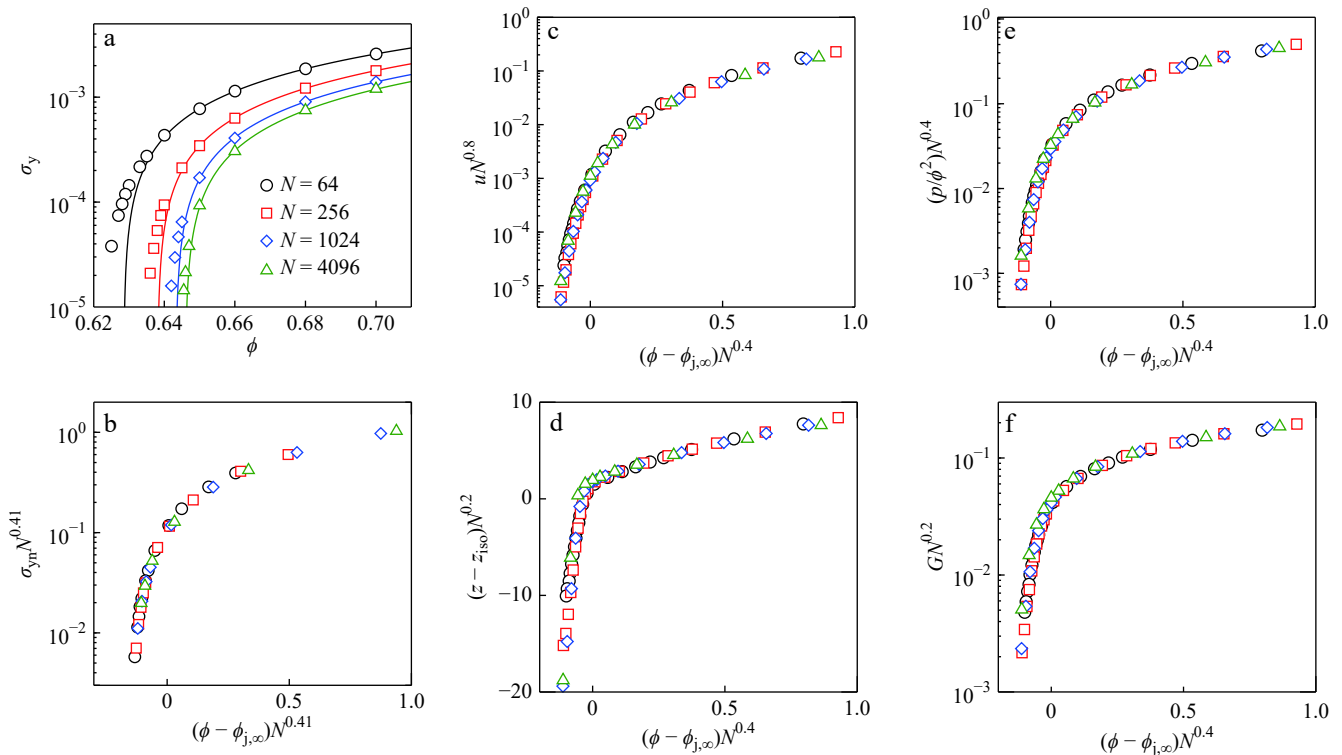


Fig. 9 (a) Yield stress σ_y against packing fraction ϕ for three-dimensional harmonic systems. The solid lines are fits to Eq. (13). (b–f) Finite size scaling of the reduced yield stress σ_{yn} , potential energy u , excess coordination number $z - z_{\text{iso}}$, pressure p , and shear modulus G , respectively. (Adapted with permission from Ref. [71]; Copyright (2014) American Physical Society)

$$\zeta \sim (\phi - \phi_{j,\infty})^{-1/3\mu} \quad (17)$$

which diverges at $\phi_{j,\infty}$.

There have been quite a few studies showing criticality of the jamming transition with diverging length scales. However, the exponent of the length scale seems not consistent in different studies, which is a puzzling aspect of the jamming transition. The robustness of the exponent obtained in our study is further strengthened by finite size scalings of other typical quantities:

$$u = U/N = (\phi - \phi_{j,\infty})^2 g_u [(\phi - \phi_{j,\infty}) N^{0.4}] \quad (18)$$

$$\frac{p}{\phi^2} = (\phi - \phi_{j,\infty}) g_p [(\phi - \phi_{j,\infty}) N^{0.4}] \quad (19)$$

$$z - z_{\text{iso}} = (\phi - \phi_{j,\infty})^{0.5} g_z [(\phi - \phi_{j,\infty}) N^{0.4}] \quad (20)$$

$$G = (\phi - \phi_{j,\infty})^{0.5} g_G [(\phi - \phi_{j,\infty}) N^{0.4}] \quad (21)$$

as shown in Figs. 9(c)–9(f). We have also verified that the exponent $\mu \approx 0.4$ holds for Hertzian systems. Therefore, our study provides strong evidence to support the criticality of the jamming transition with a well-defined scaling exponent independent of observables and interactions.

Jamming of self propelled particles

Instead of shear, a system can be driven to flow as well when particles are self propelled. As a simplified model of active matter such as birds and fish, self propelled particles (SPPs) have attracted much attention recently.^[92,93] Because of the

nonequilibrium nature, SPPs exhibit a lot of abnormal collective behaviors,^[70,94–112] which greatly enrich the contents of nonequilibrium statistical physics. In contrast to the application of shear stress which imposes a directional force parallel to the surface of the system, SSPs are propelled by random forces. Here we consider a minimal model with the following equation of motion:

$$b \frac{d\vec{r}_i}{dt} = - \sum_j \frac{dU(r_{ij})}{d\vec{r}_i} + f \vec{n}_i \quad (22)$$

where $\vec{r}_i$ is the location of particle i , b is the damping coefficient, f and $\vec{n}_i$ are the strength and orientation of self propulsion, respectively, and the sum is over all particles interacting with particle i . Although external forces are applied in different ways, intuitively, collective motion of SPPs may exhibit some similarities to sheared systems. This inspires us to study whether the criticality of the jamming transition can also be probed by SPPs.

It turns out that systems of SPPs do behave similarly to sheared systems. There is a yield self propulsion force f_y analogous to the yield stress σ_y above which the system has to flow. For two-dimensional systems with harmonic repulsion, the yield force is roughly linear well above the jamming transition,^[70] as shown in Fig. 10(a):

$$f_y(\phi, N) = k_N(\phi - \phi_{j,N}) \quad (23)$$

with

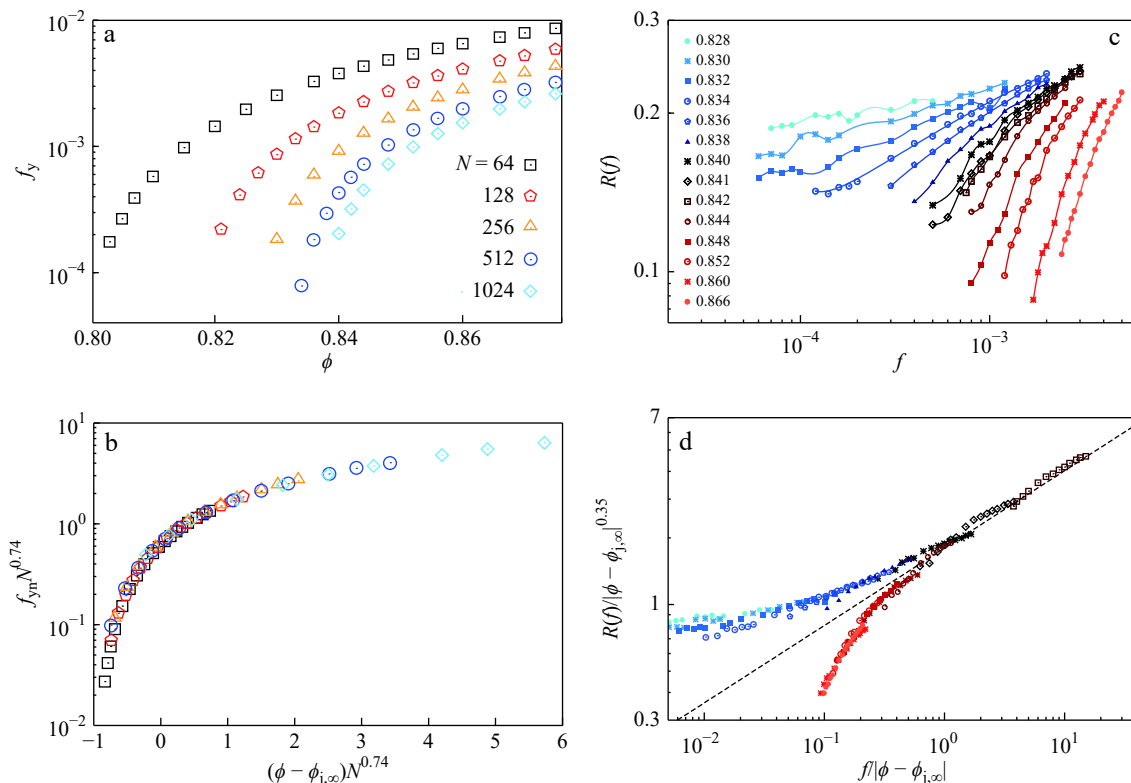


Fig. 10 (a) Yield self propulsion force f_y against packing fraction ϕ for two-dimensional harmonic systems; (b) Finite size scaling of the reduced yield force f_y ; (c) Response function $R(f)$ for various packing fractions (The lines are guides for the eye.); (d) Scaling collapse of $R(f)$ (The dashed line shows the limiting behavior (when $f/|\phi - \phi_{j,\infty}| \rightarrow \infty$) with a slope of 0.35.) (Adapted with permission from Ref. [70]; Copyright (2018) The Royal Society of Chemistry)

$$\phi_{j,N} = \phi_{j,\infty} - 0.61N^{-0.74} \quad (24)$$

$$k_N = k_\infty + 0.52N^{-0.38} \quad (25)$$

where $\phi_{j,\infty} = 0.8413 \pm 0.0002$ and $k_\infty = 0.032 \pm 0.003$ are critical packing fraction of the jamming transition and amplitude of f_y in the $N \rightarrow \infty$ limit, respectively. Note that Eqs. (23)–(25) have the same form as Eqs. (13)–(15). It is thus straightforward to expect that $f_y(\phi, N)$ may show finite size scaling as $\sigma_y(\phi, N)$ does, which is verified by Fig. 10(b). There is a master function

$$f_{yn} = \frac{f_y}{k_N} = (\phi - \phi_{j,\infty}) g_f \left[(\phi - \phi_{j,\infty}) N^{0.74} \right] \quad (26)$$

collapsing $f_y(\phi, N)$ curves for various system sizes N .

We further construct a response function $R(\phi, f) = \bar{v}(\phi)/f$ with $\bar{v}(\phi)$ being the average velocity of SPPs driven by the self propulsion force f . Interestingly, as shown in Figs. 10(c) and 10(d), there is a scaling collapse of $R(\phi, f)$ following the function:

$$R(\phi, f) = |\phi - \phi_{j,\infty}|^{0.35} h_{\pm} \left(\frac{f}{|\phi - \phi_{j,\infty}|} \right) \quad (27)$$

where h_+ and h_- are scaling functions for $\phi > \phi_{j,\infty}$ and $\phi < \phi_{j,\infty}$, respectively. Similar scaling collapse of $\eta^{-1}(\phi, \sigma_{xy})$ has been obtained for shear flows, with η being the shear viscosity.^[68]

Eqs. (26) and (27) support the criticality of the jamming transition at $\phi_{j,\infty}$ from the perspective of SPPs. Moreover, the similarities between systems of SPPs and sheared systems revealed by our work call for further investigations of the underlying connections between the two seemingly different driven systems.

Transition from Crystals to Disordered Crystals: Generalization of Jamming Physics

The jamming transition refers particularly to the formation of disordered solids with isostaticity. However, by purposely preparing the samples, *e.g.*, by decreasing the compression rate, we are able to obtain static packings of monodisperse hard spheres at packing fractions ranging from $\phi_{j,\infty}$ to ϕ_{cp} , *i.e.*, critical packing fraction of the close-packed crystalline structures such as face-centered cubic (FCC) and hexagonal close packed (HCP) in three dimensions ($d = 3$) or triangular in two dimensions ($d = 2$). Apparently, with the increase of packing fraction, the structural order increases. The states at $\phi_{j,\infty}$ correspond to the random close packing, which is the disordered end, in contrast to the other end at ϕ_{cp} with perfect structural order. It is interesting to know how a crystal evolves into a disordered solids.

A straightforward way to realize the evolution from crystals to disordered solids is to introduce and increase the particle size dispersity. Instead of studying the evolution of hard

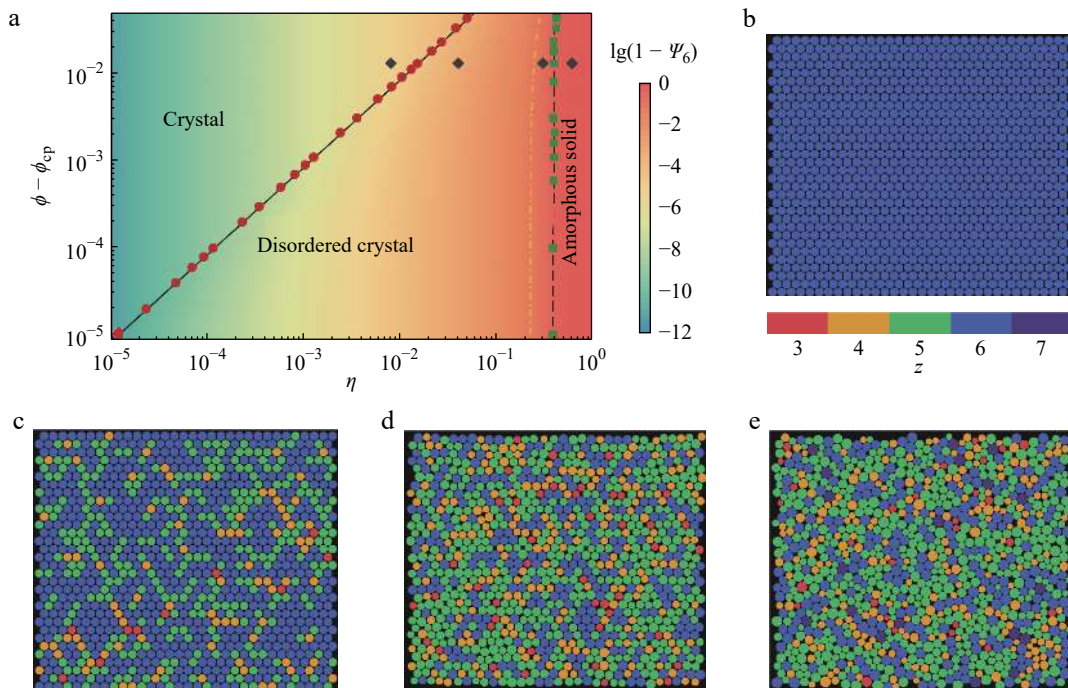


Fig. 11 (a) Phase diagram of two-dimensional harmonic systems with the transition from crystals to disordered crystals (circles) and the transition from disordered crystals to amorphous solids (squares) in the parametric space of the particle-size polydispersity η and packing fraction distance from the close packing of hard spheres $\phi - \phi_{cp}$. The solid line shows the behavior $\eta_c \sim (\phi - \phi_{cp})$ of the transition from crystals to disordered crystals. The dashed line is the guide for the eye. The color contour shows $\lg(1 - \Psi)$ with Ψ_6 being the bond orientational order. The dot-dashed line marks $\Psi_6 = 0.95$, to the left of which all states have a high crystalline order. (b–e) Configurations of a crystal, a disordered crystal, a strongly disordered crystal, and an amorphous solids whose locations in the phase diagram are labeled by the diamonds in (a). The color coding shows the coordination number of each particle. Note that without the color coding, (b) and (c) would be indistinguishable with the eyes. (Adapted with permission from Ref. [113]; Copyright (2015) Nature)

particles, here we concentrate on soft particles at $\phi > \phi_{cp}$. Starting from a packing from a close-packed structure, we continuously grow the particle size dispersity η while maintaining the packing fraction. Apparently, with sufficiently large η , the system will turn into a disordered solid.^[113,114]

To our surprise, well before the establishment of the structural disorder, there is a hidden transition from crystals to disorder crystals when η is still rather small and the crystalline order is still pretty high. Here we define disordered crystals as the solids with crystalline structure but mechanical and vibrational properties of disordered solids. As shown in Fig. 11, when η increases from 0, although the structure is still highly ordered, the force network starts to show strong spatial heterogeneity. Take two-dimensional systems as examples. When η equals 0 and the crystalline structure is perfect, all particles have 6 interacting neighbors (particle contacts). With a slight increase of η , some particles lose neighbors, leading to the spatial fluctuation of the coordination number z . We find that both the average value of z and its variance δz quickly change after a critical value of η denoted as η_c , as shown in Figs. 12(a) and 12(b). Fig. 12(c) shows that the susceptibility $\chi = N[\langle(\delta z)^2\rangle - \langle\delta z\rangle^2]$ exhibits a peak at η_c , where $\langle\cdot\rangle$ denotes the ensemble average. Interestingly, $\chi(\eta)$ curves at different packing fractions collapse nicely onto the same master curve when η is scaled by $\phi - \phi_{cp}$, as shown in Fig. 12(d). Moreover, elastic and vibrational properties undergo qualitative change across η_c . Fig. 12 shows examples of bulk modulus B , shear modulus G , compressive nonaffinity μ_c , and shear nonaffinity μ_s . All quantities remain

constant when $\eta < \eta_c$ but vary with η when $\eta > \eta_c$.

In Fig. 11(a), we plot $\phi - \phi_{cp}$ against η_c . The scaling collapse shown in Fig. 12(d) implies that $\eta_c \sim (\phi - \phi_{cp})$. Therefore, close packing at $\phi = \phi_{cp}$ is singular. We further reveal that, though highly ordered (quantified by the color contour in Fig. 11a), disordered crystals are governed by jamming physics instead of crystal physics, except for a regime right above the transition, where an intermediate physics between jamming and crystal physics exists. This greatly generalizes the jamming physics. Because strictly defined crystals only exist when the polydispersity η is small, most real systems are polydisperse and may be mainly categorized into disordered crystals. Therefore, compared to crystal physics, jamming physics covers a broader domain of materials.^[113,115]

RICH PHASE BEHAVIORS AT HIGH DENSITIES: “SOFT” ASPECT OF THE POTENTIAL

In this section, we will concentrate on high packing fractions, where the soft aspect of the potential takes effect. We will see that the soft-core nature of Eq. (1) induces some surprising phenomena and much richer phase behaviors than the hard aspect. The high packing fraction regime is where one can really appreciate the beauty of the simple potential.

Deep Jamming and Reentrant Glass Transition

In previous sections, we have discussed the scaling relations in marginally jammed solids near the jamming transition. A

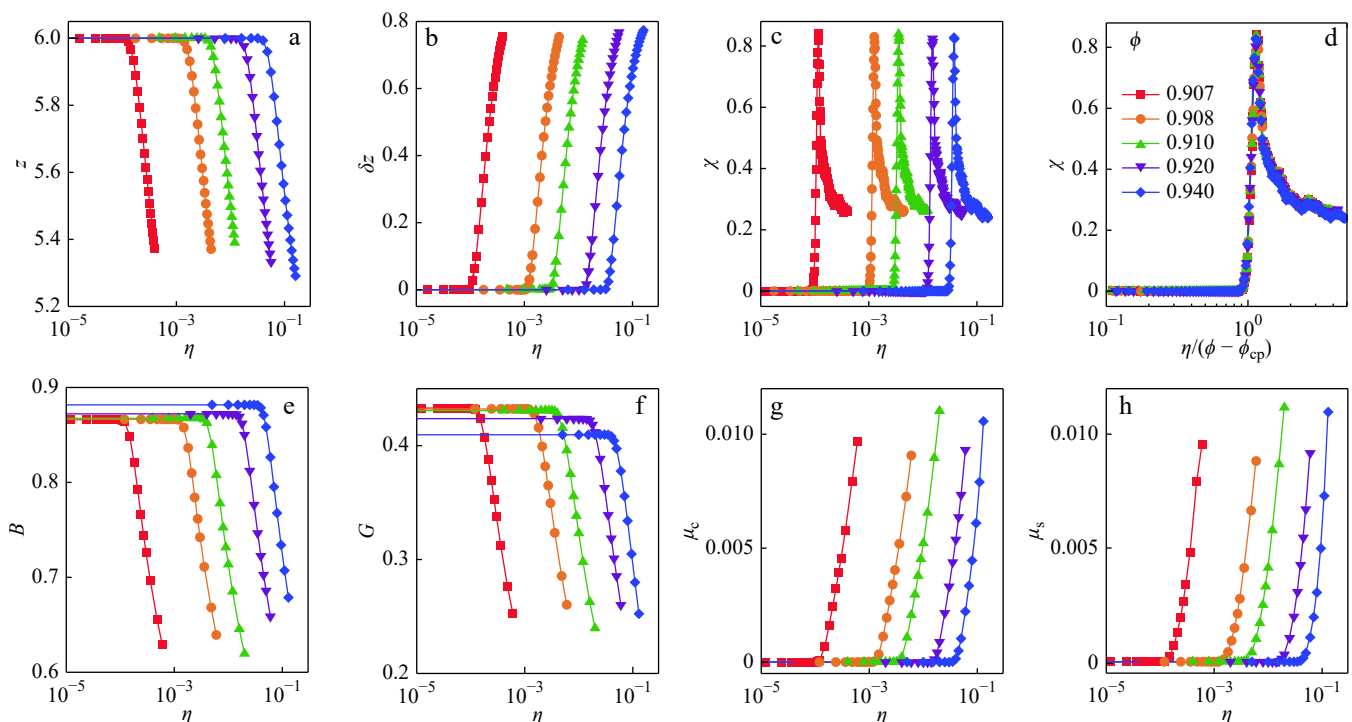


Fig. 12 (a–c) Polydispersity η evolution of the average coordination number z , fluctuation of the coordination number δz , and susceptibility χ . The critical polydispersity of the transition η_c shown in Fig. 11(a) is determined by the location of the peak in $\chi(\eta)$. (d) Scaling collapse of all curves in (c) when χ is plotted against $\eta/(\phi - \phi_{cp})$. (e–h) Polydispersity evolution of the bulk modulus B , shear modulus G , nonaffinity of the compression deformation μ_c , and nonaffinity of the shear deformation μ_s . (Adapted with permission from Ref. [113]; Copyright (2015) Nature)

question is how far away from the jamming transition can the scaling relations still hold. If the scaling relations break down at high packing fractions, are there any new relations?

In Fig. 13, we show the packing fraction evolution of potential energy U , bulk modulus B , shear modulus G , and excess coordination number beyond isostaticity $z - z_{\text{iso}}$ for both harmonic and Hertzian repulsions in three dimensions. The critical scalings of Eqs. (2)–(6) hold until $\phi = \phi_d \approx 1.18$. When $\phi > \phi_d$, Fig. 13 indicates that there is a unified scaling relation

$$\Gamma - \Gamma(\phi_d) \sim (\phi - \phi_d)^{\nu_\Gamma} \quad (28)$$

where Γ denotes U , B , G , and z . The values of ν_Γ are listed in Table 1. In contrast to Eqs. (2)–(6), here the scaling exponents ν_Γ no longer rely on potential.

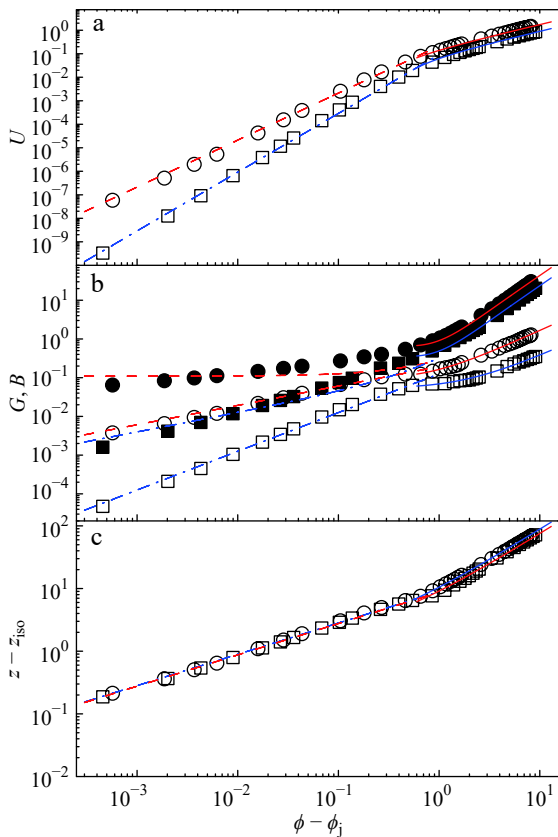


Fig. 13 (a) Potential energy U , (b) shear modulus G (empty symbols), bulk modulus B (solid symbols), and (c) excess coordination number above isostaticity $z - z_{\text{iso}}$ versus $\phi - \phi_j$ of three-dimensional jammed systems with harmonic (circles) and Hertzian (squares) interactions. The red dashed (harmonic) and blue dot-dashed (Hertzian) lines are fits for marginally jammed solids according to Eqs. (2), (4)–(6), while the solid curves are fits for deeply jammed solids according to Eq. (28). (Adapted with permission from Ref. [116]; Copyright (2011) American Physical Society)

Table 1 Exponents ν_Γ in Eq. (28) for both harmonic and Hertzian systems

α	ν_U	ν_B	ν_G	ν_z
2	1.02 ± 0.02	1.71 ± 0.02	1.16 ± 0.03	0.97 ± 0.03
$\frac{5}{2}$	1.03 ± 0.02	1.73 ± 0.03	1.19 ± 0.05	1.03 ± 0.02

We call jammed states at $\phi > \phi_d$ as deeply jammed states.^[116] Therefore, $\phi = \phi_d$ is the transition point from marginal jamming to deep jamming, where jammed solids undergo qualitative changes in material properties.

We further notice that the stability of jammed solids undergoes a nonmonotonic change at ϕ_d . Jammed solids are most stable at ϕ_d , which have the highest energy barriers and weakest localization of the low-frequency modes of vibration. From the perspective of solids, the glass transition temperature reflects the stability of disordered solids.^[117,118] It is thus expected that the glass transition temperature of the model systems studied here should reach the maximum around ϕ_d . This is verified by our simulations. Like the reentrant crystallization shown in Fig. 5, the glass transition temperature is reentrant, with the maximum being roughly at $\phi = \phi_d$.^[119]

The reentrant crystallization and glass transition are unique to soft-core potentials. Because of the reentrance, soft-core particles can form various types of solids. Moreover, the opposite pressure (or density) dependence of the crystallization (melting) or glass transition temperature on both sides of the maximum implies the possibility to observe different behaviors in the liquid-solid transitions. In the following, we will show two unexpected phenomena resulted from the reentrant crystallization.

Discontinuous to Continuous Hexatic-liquid Transition in Two-dimensional Melting

As mentioned in Sec. I, it has been settled that the hexatic-liquid transition in two-dimensional melting of hard particles is discontinuous.^[24] It has also been shown that with the increase of particle softness, such a discontinuity in systems with an inverse power law interaction is weaker and the transition eventually becomes continuous when particles are soft enough.^[26] Therefore, the hard and soft dual nature makes soft-core particles desirable in the study of two-dimensional melting. Note that here the particle softness is not tuned. The particles are intrinsically soft. Only that the density or pressure controls the particle overlap and thus acts as the driving force to effectively “tune” the softness. Moreover, inverse power law potential is unbounded, which is intrinsically different from the soft-core potentials. It is thus interesting to know how the hexatic-liquid transition of soft-core particles evolves with density and whether the reentrant crystallization (melting) has any consequence on the nature of the transition.

At fixed temperature below the maximum melting temperature T_m (occurring at a density ρ_m), we carefully identify states across the melting line for monodisperse soft-core particles. In two dimensions, solids have long range bond orientational order (BOO) and quasi long range positional order (PO). Both orders are short ranged in liquids. Hexatic phase is in between, which has quasi long range BOO and short range PO. Therefore, all three phases can be identified by the spatial correlation functions of BOO and PO.^[11,120]

In Fig. 14(a), we show the equation of state of Hertzian systems at fixed temperature on the lower density side of ρ_m . There is clearly a Mayer-Wood loop, signaling the phase coexistence and the first order nature of the transition. We veri-

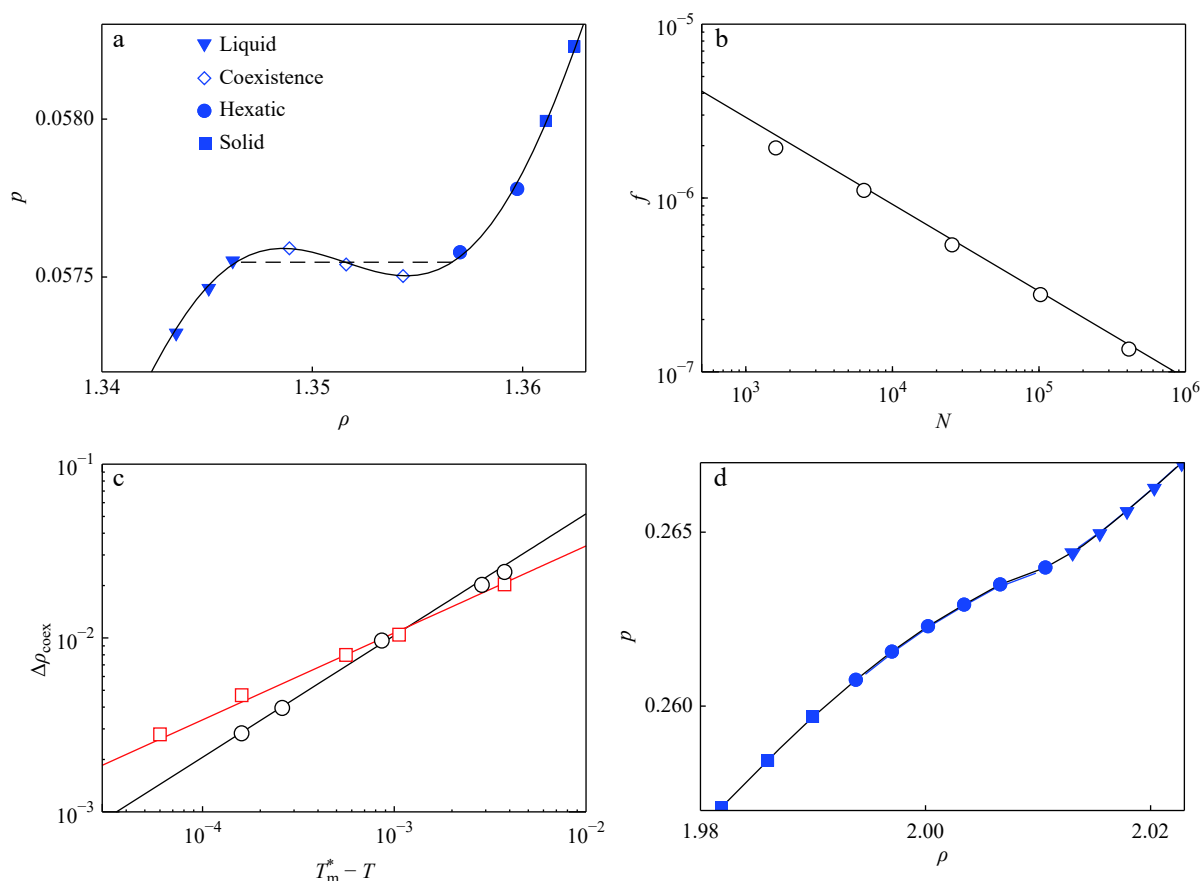


Fig. 14 (a) Isothermal equation of state $p(\rho)$ calculated at a fixed temperature across the melting at $\rho < \rho_m$ for $N = 102400$ Herztian particles in two dimensions. The solid line is a tenth-order polynomial fit to the data. The dashed line demonstrates the Maxwell construction. (b) System size dependence of the interface free energy per particle f calculated from the Mayer-Wood loop in (a). The line shows the scaling: $f \sim N^{-1/2}$. (c) Temperature dependence of the density interval of phase coexistence $\Delta\rho_{\text{coex}}$ for Herztian (circles) and harmonic (squares) repulsions. The lines show the scaling: $\Delta\rho_{\text{coex}} \sim (T_m^* - T)^s$, with T_m^* being approximately the maximum melting temperature T_m . (d) Isothermal equation of state $p(\rho)$ calculated for the same system and at the same temperature as (a), but across the transitions at $\rho > \rho_m$. The line is a guide for the eye. (Adapted with permission from Ref. [28]; Copyright (2016) American Physical Society)

fy that liquid and hexatic phases indeed coexist in the loop. The area encircled by the loop and the Maxwell construction (dashed line in Fig. 14a) sets the interface free energy per particle f between the coexistent phases. Fig. 14(b) shows $f \sim N^{-1/2}$, which is another evidence of the first order transition from hexatic phase to liquids.

Fig. 14(c) shows that the density interval of the coexistence, $\Delta\rho_{\text{coex}}$, decreases with increasing temperature according to the power law: $\Delta\rho_{\text{coex}} \sim (T_m^* - T)^s$, where s and T_m^* are fitting parameters. Interestingly, T_m^* is approximately equal to the maximum melting temperature T_m . Therefore, approaching T_m , $\Delta\rho_{\text{coex}}$ decays to zero and the hexatic-liquid transition becomes continuous.

When $\rho > \rho_m$, there is no longer the Mayer-Wood loop in the equation of state. As shown in Fig. 14(d), the pressure increases monotonically with density. Therefore, on the higher density side of the maximum melting temperature, two-dimensional melting is always continuous. The state point at (ρ_m, T_m) acts as the transition point between discontinuous and continuous hexatic-liquid transitions.

We further find that the same scenarios occur in other

soft-core particle systems, e.g., Gaussian-core particles. Therefore, the reentrant crystallization has strong correspondence with the nature of two-dimensional melting, which should be a general feature of soft-core particle systems.

Unexpected Formation of Quasicrystals

A fantastic part of the soft-core model of Eq. (1) is that with the increase of density (or packing fraction) there are multiple reentrant crystallizations.^[121] Each reentrance corresponds to a ground state with a particular structure. In Fig. 5, we only show the first two reentrances for two-dimensional Herztian systems with triangular and square lattice structures, respectively. In Fig. 15, we show the phase diagram of solid states in two dimensions over a wide range of densities, together with the configurations and diffraction patterns for periodic crystals found here. It is intriguing that such a simple soft-core system can form so many solid structures, including triangle, square, honeycomb, kite, sigma phase, string, and rhombus. There are more structures when the exponent of the soft-core potential α decreases. The most striking and surprising finding is that quasicrystals exist with

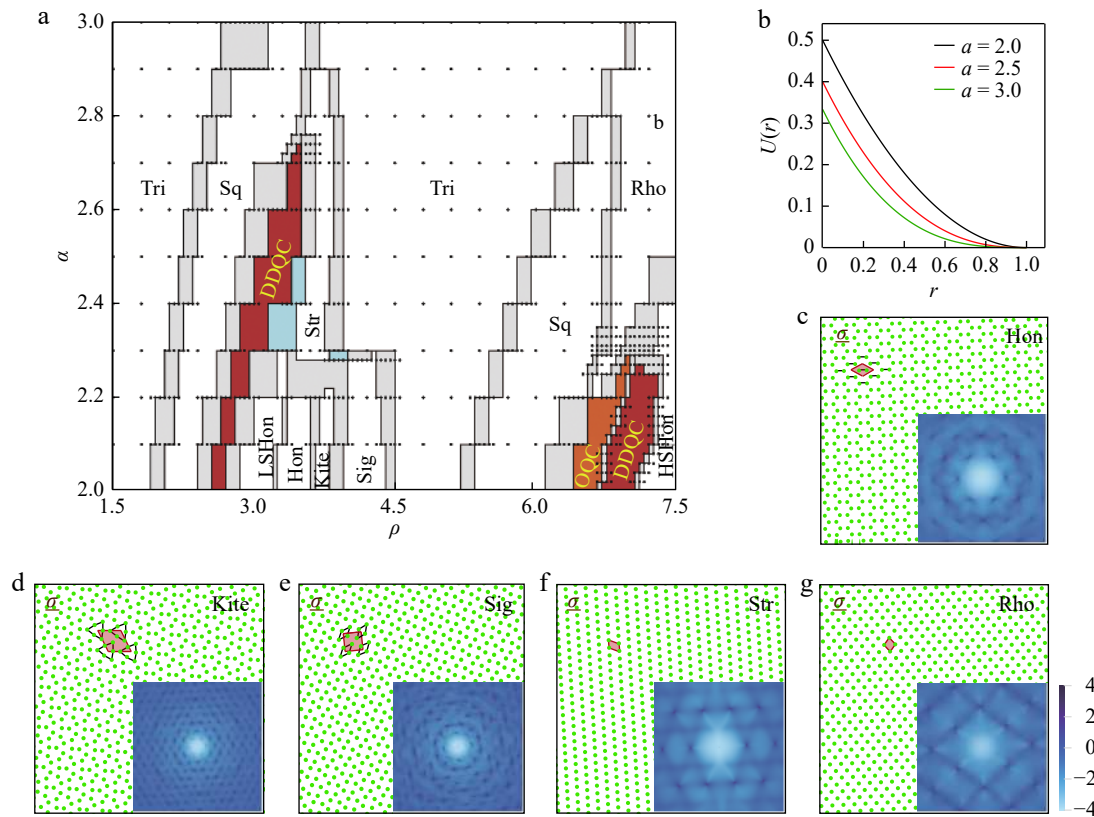


Fig. 15 (a) Phase diagram of solid states in terms of number density ρ and potential exponent α in two dimensions. The black dots label the (ρ, α) pairs where we run simulations to identify states. The orange and red areas are the territories of OQCs and DDQCs, respectively. The gray and light-blue areas are phase coexistence regimes of two and three types of solids, respectively. Tri, Sq, Str, Hon, Kite, Sig, Rho, LSHon, and HSHon denote triangle, square, string, honeycomb, kite, sigma phase, rhombus, lower-density stretched honeycomb, and higher-density stretched honeycomb, respectively. (b) Examples of particle interaction potentials with different α (Eq. 1). (c–g) A part of static configurations of five distinct crystalline solids with the scale bar indicating the actual size of particle diameter σ . The red-shaded polygons and black lines outline the unit cell and basis, respectively. The insets are diffraction patterns with the values of the scale bar being in the logarithmic scale. (Adapted with permission from Ref. [122]; Copyright (2017) Nature)

certain values of α and ρ .^[122] As shown in Fig. 15, we find both dodecagonal and octagonal quasicrystals (DDQCs and OQCs). Note that prior to us, there has been no convincing report of the finding of OQCs in soft matter systems.

From previous studies of quasicrystals, there seems to be a consensus that multiple length scales to stabilize quasicrystals need preparing in advance. As mentioned earlier, one can use bidisperse particles (*e.g.*, alloys), anisotropic particles (*e.g.*, patchy or tetrahedron particles), or monodisperse and isotropic particles with a special interaction containing multiple length scales. The most surprising aspect of the quasicrystals found in our work is that no multiple length scales are purposely prepared. We use monodisperse particles with isotropic interactions. As shown in Fig. 15(b), the interaction potentials are smooth without any explicit length scales. Therefore, the finding of such quasicrystals violates the prevailing two-length scenario of the quasicrystal formation.^[36] The complicated interplay between high density and soft nature of the potential is the key to understanding the quasicrystal formation, which calls for future studies.

A unique feature of our quasicrystals is that particles tend to form pentagons, which are building blocks of the quasi-

crystals. Fig. 16 shows the structural and dynamical characterizations of both OQCs and DDQCs. Pentagons are everywhere. When the centers of adjacent pentagons are connected, the whole system is divided into a beautiful tiling of polygons (square-rhombus in OQCs and square-triangle in DDQCs). The 8- and 12-fold rotational symmetries of OQCs and DDQCs can be clearly seen from the static structure factor $S(\vec{k}) = N^{-1} \langle \rho(\vec{k}) \rho(-\vec{k}) \rangle$, pair correlation function $g(\vec{r}) = 2^{-1} L^2 N^{-2} \langle \sum_{i=1}^N \sum_{j \neq i}^N \delta(\vec{r} - \vec{r}_{ij}) \rangle$, and van Hove autocorrelation function $G_a(\vec{r}, t) = \langle N^{-1} \sum_{i=1}^N \delta[\vec{r} - \vec{r}_i(t) + \vec{r}_i(0)] \rangle$, where $\rho(\vec{k}) = \sum_{j=1}^N e^{i\vec{k} \cdot \vec{r}_j}$ is the Fourier transform of the density with $\vec{k}$ being the wave vector satisfying the periodic boundary conditions, and $\langle \cdot \rangle$ denotes the ensemble average. The importance of the pentagons can also be seen from the particle trajectories shown in Figs. 16(d) and 16(i). A particle moves along the edge of a pentagon for sometime and then suddenly jumps out and forms another pentagon with new neighbors, corresponding to a phason flip. Because of phason flips, the long time dynamics of the quasicrystals is diffusive.^[123–125]

The mystery of the quasicrystal formation in our soft-

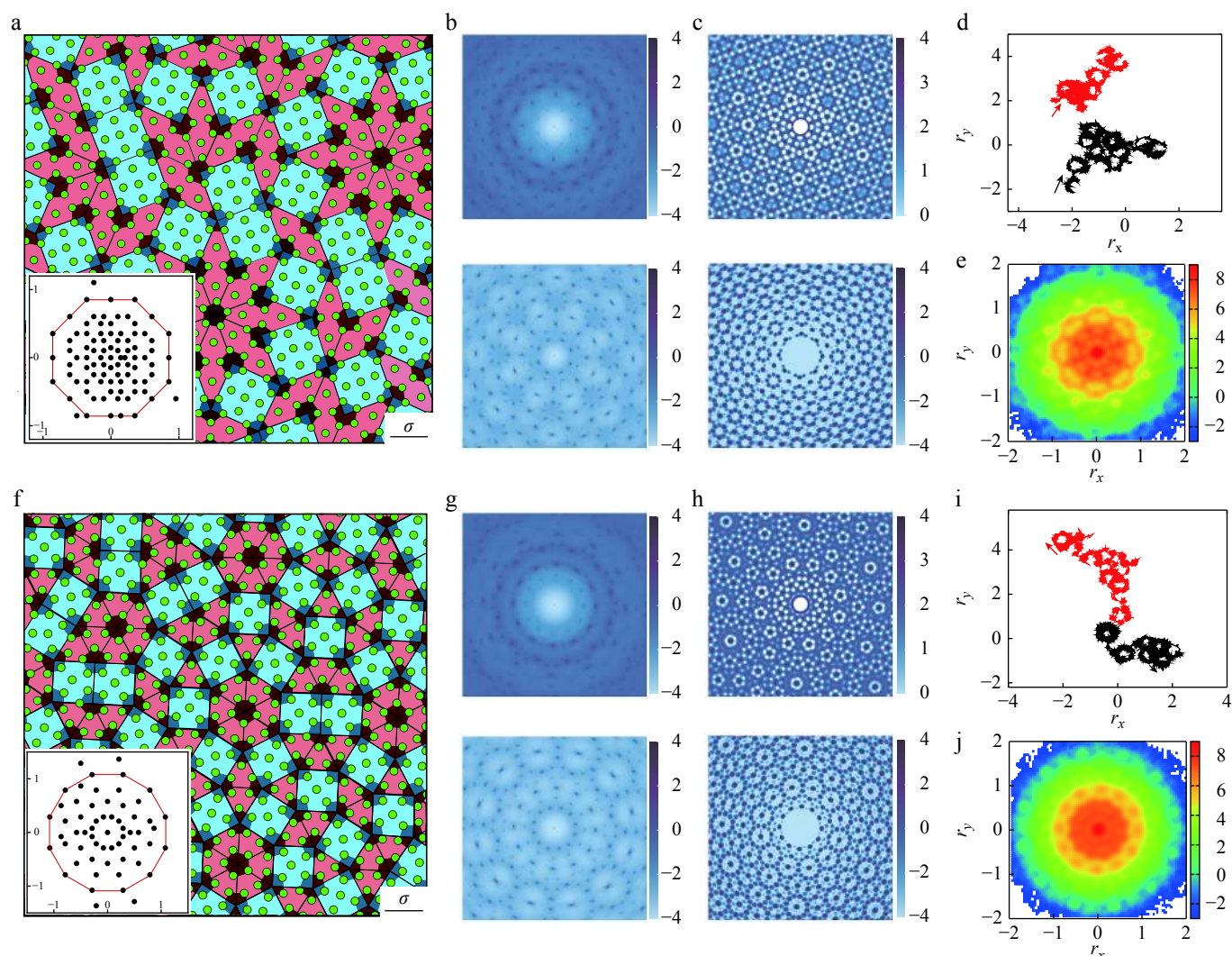


Fig. 16 (a–e) An OQC and (f–j) a DDQC at $(\rho, \alpha, T) = (6.6, 2.0, 1.36 \times 10^{-3})$ and $(7.0, 2.0, 1.72 \times 10^{-3})$, respectively: (a, f) a part of static configuration with the square-rhombus (square-triangle) tiling (The black scale bar indicates the actual size of particle diameter σ . The tessellation is constructed by connecting the centers of pentagons. The inset shows the projection of quasicrystal in the perpendicular space with the red polygon being the atomic surface.); (b, c, g, h) Diffraction patterns and density profiles calculated from single particles (top panel) and from pentagons (bottom panel); (d, i) Particle trajectories with the arrows pointing to the original direction of motion; (e, j) van Hove autocorrelation function $G_a(\vec{r}, t)$ at a instant $t = 6000$ (The values of the scale bars are in the logarithmic scale.) (Adapted with permission from Ref. [122]; Copyright (2017) Nature)

core particle systems is how the length scales stabilizing the quasicrystals spontaneously emerge. The key is why particles tend to form pentagons. More studies are required to reveal the underlying mechanisms of such a quasicrystal formation, in perspective of both thermodynamics and kinetic pathways.

CONCLUSIONS AND OUTLOOK

In this paper, we review some of our recent progress in the study of phase behaviors in a very simple soft-core model system. Prior to us, it has already been known that such a simple system can form multiple solid phases with the increase of density.^[61] With extra carefulness, we are able to move forward to reveal some hidden phenomena. The emergence of rich phases makes the model system ideal to study the fundamental problems of phase transitions and self

assembling, which are also highly regarded issues in polymer science. We have just moved a small step without a full understanding of the findings. There are thus many remaining and new issues to be attacked in future studies.

How to verify our findings in realistic systems is apparently a problem to be settled. As mentioned earlier, some experimental systems such as PNIPAM microgels exhibit interactions similar to Hertzian repulsion. Therefore, PNIPAM microgels have been successfully applied in the experimental studies of the jamming transition and other phase transitions, *e.g.*, two-dimensional melting. However, PNIPAM microgels cannot be squeezed into so high densities as in our simulations, so it remains rather difficult to find or fabricate an experimental system which can resemble our soft-core potentials and enter the high density regime. Star polymers^[126] or paramagnetic colloids with long range interac-

tions^[127] are possible candidates, which may be used to study two-dimensional melting or reentrant crystallization. However, the formation of quasicrystals is sensitive to the actual form of the interaction, as illustrated by Fig. 15, which requires subtle fabrications of colloidal suspensions.

Follow-up studies are required to forward our understanding of the problems concerned in our work. Although significant advances have been achieved in the understanding of the jamming transition, we are still on the way of revealing its true nature. Among many questions about the jamming transition, we are specially interested in building up a bridge between disordered solids and crystals. We expect that such a bridge may facilitate the revelation of the nature of disordered solids, from the evolutionary perspective of crystals. For two-dimensional melting and quasicrystals, we just report the phenomena. More work is definitely required to reveal the underlying mechanisms of these novel findings. Moreover, there are many different crystals and quasicrystals adjacent to each other. Each solid phase corresponds to a reentrant crystallization. It is interesting to know whether the two-stage scenario of two-dimensional melting can be extended to other solid symmetries than triangle and whether there is still a discontinuous-continuous transition. The adjacency of different types of solids is also ideal to study solid-solid transitions.^[128]

In the studies shown here, we only concern about phase transitions beyond the glass transition density of repulsive systems. If attraction is present, we are able to see the formation of solid-like states such as gels or attractive glasses at lower densities and revisit the jamming scenario from the perspective of weakly attractive particulate systems.^[129–132] There remain lots of interesting phenomena and mechanisms of phase transitions to explore for the sticky matter at low densities. The experimental realization of colloidal suspensions with a tunable attraction and synthesis of special sticky particles such as patchy colloids are necessary in this set of studies, which require the deep involvement and application of polymer science.

ACKNOWLEDGMENTS

We would like to thank all the collaborators in all the studies presented in this paper and the support from the National Natural Science Foundation of China (Nos. 11734014, 11574278, 21325418, 11074228, and 91027001), the National Basic Research Program of China (973 Program) (No. 2012CB821500), CAS 100-Talent Program (No. 2030020004), and Fundamental Research Funds for the Central Universities (Nos. 2340000034 and 2030020028).

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