Shear-induced solidification of athermal systems with weak attraction

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We find that unjammed packings of frictionless particles with rather weak attraction can always be driven into solidlike states by shear. The structure of shear-driven solids evolves continuously with packing fraction from gel-like to jamminglike, but is almost independent of the shear stress. In contrast, both the density of vibrational states (DOVS) and force network evolve progressively with the shear stress. There exists a packing fraction independent shear stress \( \sigma_c \), at which the shear-driven solids are isostatic and have a flattened DOVS. Solidlike states induced by a shear stress greater than \( \sigma_c \) possess properties of marginally jammed solids and are thus strictly defined shear jammed states. Below \( \sigma_c \), shear-driven solids with rather different structures are all under isostaticity and share common features in the DOVS and force network. Our study leads to a jamming phase diagram for weakly attractive particles, which reveals the significance of the shear stress in determining properties of shear-driven solids and the connections and distinctions between jamminglike and gel-like states.

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I. INTRODUCTION

Particulate systems such as colloids, emulsions, foams, and granular materials can form disordered solids at high packing fractions [1–11]. The critical packing fraction of the transition from liquidlike to solidlike states is sensitive to the interaction [11–13] and geometry [14–17] of particles. Consider the simplest case of static packings of spheres. If the spheres are frictionless and purely repulsive, the transition happens as the jamming transition at a critical packing fraction \( \phi_j \), which remains rather poor over a surprisingly long time, while such knowledge is important in practice because many real systems unavoidably contain cohesive particles. Up to date, what we can learn about the jamming of attractive particles may be still from an early experimental study [3]. That work shows expectable results that the presence of attraction smears out \( \phi_j \), so that a nonvanishing yield stress or glass transition temperature extends to low packing fractions. However, there are in-depth questions that ought to be but have not yet been attacked. For instance, in the zero attraction limit, will attractive particles remain rather poor over a surprisingly long time, while such knowledge is important in practice because many real systems

\[ U(r_{ij}) = \begin{cases} \frac{\xi}{\sigma} \left( \frac{1}{2} - \frac{\rho_i}{\sigma d_{ij}} \right) \left( 1 - \frac{\rho_i}{\sigma d_{ij}} \right), & \frac{\rho_i}{\sigma d_{ij}} \leq 1 + \mu, \\ -\frac{\xi}{\sigma} \left( 1 + 2 \mu - \frac{\rho_i}{\sigma d_{ij}} \right), & 1 + \mu < \frac{\rho_i}{\sigma d_{ij}} \leq 1 + 2 \mu, \\ 0, & \frac{\rho_i}{\sigma d_{ij}} > 1 + 2 \mu, \end{cases} \]  

where \( r_{ij} \) and \( d_{ij} \) are the separation between particles \( i \) and \( j \) and sum of their radii, and \( \mu \) is a tunable parameter to control the range and strength of attraction. We study both harmonic \((\alpha = 2)\) and Hertzian \((\alpha = 5/2)\) systems. We set the units of...
mass, energy, and length to be particle mass \( m \), characteristic energy scale of the potential \( \epsilon \), and small particle diameter \( d_s \).

The shear deformation is realized by introducing the shear strain \( \gamma \) and applying the Lees-Edwards boundary conditions [29]. Without shear (\( \gamma = 0 \)), we generate static states at fixed packing fraction by applying the fast inertial relaxation engine (FIRE) method [30] to minimize the potential energy \( U = \sum_{ij} U(r_{ij}) \) of random configurations, where the sum is over all pairs of particles. To perform quasistatic shear, we successively increase the shear strain \( \gamma \) by a step strain \( \Delta \gamma \), followed by the minimization of \( U \). To well control the shear stress \( \sigma \), we instead minimize a thermodynamiclike potential \( H = U - \sigma \gamma L^d \) [24], where \( d \) is the dimension of space. During the minimization, \( \sigma \) is fixed, while \( \gamma \) becomes a variable.

The structure of the system is characterized by the static structure factor \( S(q) = \langle \sum_j \exp(iq \cdot \vec{r}_j) \rangle^2/N \), where \( q = |\vec{q}| \) is the angular wave number, \( \vec{r}_j \) is the location of particle \( j \), \( \langle \cdot \rangle \) denotes the average over states, and the sum is over all particles. The normal modes of vibration are obtained from diagonalizing the Hessian matrix using ARPACK [31]. The DOVS is then calculated as \( D(\omega) = \langle \sum_j \delta(\omega - \omega_l) \rangle/dN \), where \( \omega_l \) is the frequency of the \( l \)th normal mode of vibration, \( \langle \cdot \rangle \) denotes the average over configurations, and the sum is over all modes.

III. RESULTS

In this work, we are mainly concerned about the solidification in the \( \sigma - \phi \) plane at \( T = 0 \). If not specified, results are shown for \( N = 1024 \) harmonic systems in two dimensions. We will also show (and specify) results of system size effects, Hertzian systems, and three-dimensional harmonic systems to generalize our major findings.

A. Shear-induced solidification

We vary the strength of attraction \( \mu \) from \( 10^{-2} \) to \( 10^{-6} \), approaching the zero attraction limit. Figure 1(a) shows the probability \( P_0(\phi) \) of finding solidlike states for different values of \( \mu \) by quickly minimizing the potential energy of 10 000 random states without shear. With increasing \( \mu \), \( P_0(\phi) \) shifts to lower packing fractions. Employing the definition in early work [5], we determine the jamming transition threshold \( \phi_{j,\mu} \) from \( P_0(\phi) \). As shown in Fig. 1(b), \( \phi_j - \phi_{j,\mu} \sim \mu^{1/3} \) in the small \( \mu \) limit, where \( \phi_j \approx 0.842 \). Without shear, the attraction seems to play the role of a perturbation.

Figure 1(a) indicates that well below \( \phi_{j,\mu} \), by the direct-queench sampling without shear, it is almost impossible to find solidlike states. Starting from unjammed states, we apply quasistatic shear. As shown in Fig. 1(c), in the early stage of the quasistatic shear, the system remains unjammed (\( \sigma = 0 \)). Interestingly, as long as the step strain \( \Delta \gamma \) is small enough, nonzero shear stress emerges and fluctuates when \( \gamma > \gamma_c \), signaling the formation of solidlike states. The solid forming ability under quasistatic shear decreases when packing fraction decreases, demonstrated by the growth of \( \langle \gamma_c \rangle \) in Fig. 1(d), where \( \langle \cdot \rangle \) denotes the average over independent runs of quasistatic shear.

A similar phenomenon called shear jamming has also been observed in packings of frictional particles [11]. For frictional particles, jamming can happen at packing fractions lower than \( \phi_j [13,32] \), where both jammed and unjammed states exist. Consequently, originally unjammed packings of frictional spheres can jam under quasistatic shear [11]. The underlying physics of such a shear jamming phenomenon still remains elusive and is a hot topic in the field of granular materials [33,34]. In comparison, the shear-induced solidification of attractive particles illustrated in Fig. 1 may convey richer and more complicated physics, at least because of the possible existence of various types of solidlike states. Furthermore, attraction and friction are intrinsically distinct in stabilizing particles. It is interesting to know how attractive particles behave under shear and whether their behaviors could also shed some light on our understanding of shear jamming of frictional particles.

B. Yield stress

During quasistatic shear, the shear stress fluctuates and is not controllable, as shown in Fig. 1(c). To have a clear picture of the shear stress dependence (which turns out to be important), we instead sample shear-driven solids by a recently developed algorithm [23,24] to well control the shear stress. By minimizing a thermodynamiclike potential \( H \) of random states, as introduced in Sec. II, we can quickly look for solidlike states at desired shear stress. As shown in Fig. 1, it takes some shear strain to find shear-driven solids. Furthermore, it is impractical to let \( \gamma \to \infty \). We thus set a maximum strain \( \gamma_m = 20 \). The search for shear-driven solids fails once \( \gamma > \gamma_m \).

For each pair of \( \phi \) and \( \sigma \), we run 1000 independent trials and calculate the probability of finding solidlike states \( P(\sigma, \phi) \), as shown in Fig. 2(a). At fixed \( \phi \), \( P(\sigma) \approx 1 \) when \( \sigma \) is small. For finite size systems, \( P(\sigma) \) decreases with increasing \( \sigma \) near the yield stress \( \sigma_y [23,24,35] \), which we determine from...
$P(\sigma_i) = 0.5$. Figure 2(b) shows $\sigma_i(\phi)$ for different strength of attraction $\mu$. $\sigma_i$ decreases with decreasing $\phi$, and remains nonzero down to rather low packing fractions. At fixed $\phi$, $\sigma_i$ decreases when $\mu$ decreases. Interestingly, roughly below the rigidity percolation threshold at $\phi_{rp}\approx 0.689$ [12], $\sigma_i \sim \mu$, while it breaks down when $\phi > \phi_{rp}$. We will see later that $\phi_{rp}$ does not happen to do so. Figure 2(b) indicates that in the small attraction limit the attraction does not act as a perturbation [36–40] in the presence of shear. As will be shown, the attraction always induces multiple types of solidlike states far below the jamming threshold and qualitatively alters the jamming phase diagram for purely repulsive particles.

Systems at $\phi > \phi_{j,\mu}$ are essentially jammed without the need of shear, which does not interest us here. The focus of this work is on the regime of $\phi < \phi_{j,\mu}$, where direct quenching always finds unjammed states and solidlike states can be explored with the help of shear. In the following, we will mainly discuss shear-driven solids at $\phi < \phi_{j,\mu}$. Results for $\phi > \phi_{j,\mu}$ are presented just for comparison.

C. “Time step” issue of the minimization and protocol dependence

The FIRE method that we use to find solid-like states is based on molecular dynamics simulation, which involves a tunable “time step” $\delta t$ [30]. Because the strength of attraction $\mu$ that we study is so small, the preset $\delta t$ has to be small enough to ensure small enough step increases of shear strain. Large step increases of shear strain induced by a large preset $\delta t$ will break bonds and lead to the failure of finding shear-driven solids. As shown in Fig. 3(a), at fixed packing fraction, the probability of finding shear-driven solids $P(\sigma)$ under applied shear stress $\sigma$ increases with decreasing the initial input value of $\delta t$ as expected and becomes saturate when $\delta t$ is small enough. We always use small enough $\delta t$ when calculating $P(\sigma)$.

It has been known that the critical packing fraction $\phi_j$ of the jamming transition for purely repulsive particles depends on protocols [5,18–22]. For instance, $\phi_j$ could vary with the rate of compression and initial states. A relatively unbiased way to locate $\phi_j$ is to quickly quench ideal gas states to local potential energy minima and calculate the probability of finding jammed states at fixed packing fraction, as discussed in Ref. [5] and in Fig. 1(a). This protocol unbiasedly samples the inherent structures [19], which do not encounter the compression rate or route dependence.

In this work, we employ the same protocol to sample shear-driven solids at fixed packing fraction $\phi$ and shear stress $\sigma$, by minimizing the thermodynamiclike potential $H$. We estimate the probability of finding shear-driven solids $P(\sigma)$ and determine the yield stress from it. Therefore, each data point on the yield stress curve $\sigma(\phi)$ shown in Fig. 2(b) results from the direct sampling of the configurational space under the constraint of constant packing fraction and shear stress. It has nothing to do with the route approaching yielding, e.g., along constant packing fraction or along constant shear stress.

We have to acknowledge that, if we approach the yield stress by successively compressing (decompressing) states at fixed shear stress or decreasing (increasing) shear stress at fixed packing fraction, the yield stress may show route dependence. This route dependence should be a quite interesting issue to attack, which is out of the scope of current study. We hope to investigate it in detail in follow-up studies. The presence of attraction may lead to more complicated protocol dependence than purely repulsive systems. As demonstrated in Fig. 3(b), under quasistatic shear, an initially unjammed state with attraction evolves into solidlike states when the shear strain $\gamma$ is greater than the threshold value $\gamma_c$. When we quasistatically shear the system backwards, the system cannot return to the unjammed state, but remains rigid. This hysteresis results from the presence of attraction and implies the possible complexity of the protocol dependence, which has also been observed in the same system under compression and decompression [12].

D. Shear stress and packing fraction dependence

Figures 4(a)–4(f) are configurations with force network of shear-driven solids at different packing fractions and shear stresses. At $\phi \ll \phi_{j,\mu}$, attraction (red bonds) dominates and the states look gel-like with fractal structures. Slightly above $\phi_{j,\mu}$, the structure looks uniform and particle interactions are predominantly repulsive (blue bonds). In between, with
increasing packing fraction, the structure evolves from gel-like to jamminglike.

Figures 4(g)–4(i) demonstrate the packing fraction and shear stress evolution of the static structure factor $S(q)$. The structure is almost independent of shear stress, while it evolves strongly with packing fraction. At $\phi \ll \phi_{j,\mu}$, the low $q$ part of $S(q)$ exhibits the typical gel-like feature $S(q) \sim q^{-d_f}$ with $d_f \leq 2$ being the fractal dimension [41]. The low $q$ part of $S(q)$ moves down with increasing packing fraction, and eventually becomes flat (jamminglike feature [42–44]) near $\phi_{j,\mu}$.

Purely from the packing fraction evolution of $S(q)$, we cannot determine the boundary between gel-like and glass or jamminglike states. Note that the solidlike states are shear induced. Although the structure is insensitive to the change of shear stress, other quantities may exhibit shear stress dependence and provide useful information to distinguish states. Comparing states at the same packing fraction but different shear stresses [e.g., Figs. 4(b) and 4(e)], we can tell that the shear stress indeed remarkably affects the force network: More particles interact and repulsion plays a more important role with increasing shear stress.

Resulting from significant changes in the force network, vibrational properties of shear-driven solids exhibit strong shear stress dependence. Figures 4(j)–4(l) show the shear stress evolution of the DOVS, $D(\omega)$. When $\phi > \phi_{j,\mu}$, applying shear stress only weakly affects the force network and elastic properties [23,24]. As shown in Fig. 4(l), $D(\omega)$ at different shear stresses overlap. Interestingly, Figs. 4(j) and 4(k) show that $D(\omega)$ has strong shear stress dependence when $\phi < \phi_{j,\mu}$. For solidlike states induced by small shear stresses, there is a low-frequency peak in $D(\omega)$, indicating the aggregation of soft modes. With increasing shear stress, the peak moves down and to higher frequencies, implying the decrease of the amount of soft modes and that shear-driven solids become stiffer and more stable.

At all packing fractions below $\phi_{j,\mu}$, the motion of the low-frequency peak in $D(\omega)$ with the change of shear stress follows the same trend. However, at low packing fractions where shear-driven solids are typically gel-like, until at the yield stress, the peak is still present. In contrast, near $\phi_{j,\mu}$, the peak disappears at a crossover shear stress $\sigma_c < \sigma_y$.

Meanwhile, $D(\omega)$ exhibits a plateau, which is actually one of the most representative features of marginally jammed solids of purely repulsive particles [25–27,45]. When $\sigma > \sigma_c$, the evolution of $D(\omega)$ looks like that of marginally jammed solids under compression, but here the shear stress is the driving force instead of the packing fraction.

For marginally jammed solids, the flattening of $D(\omega)$ is associated with isostaticity [25–27,45], i.e., the average coordination number $z = z_c = 2d$ with $d$ being the dimension of space. Is the emergence of the plateau in $D(\omega)$ at $\sigma_c$ also related to isostaticity?
E. Isotaticity and strictly defined shear jamming

Figure 5(a) shows the shear stress evolution of the average coordination number (rattlers excluded) at $\phi < \phi_{j,\mu}$. When the shear stress is small, there is a plateau in $z(\sigma)$. The plateau value approaches $z_c$ from below with increasing packing fraction. Approaching the yield stress, the coordination number grows quickly and collapses onto a master curve. In each $z(\sigma)$ curve, data points at $\sigma < \sigma_c$ and with the low-frequency peak in $D(\omega)$ are denoted by blue symbols, while red symbols represent data at $\sigma > \sigma_c$. Surprisingly, $z = z_c = 4$ is exactly the boundary between two colors, indicating that isotaticity is indeed coupled to the flattening of $D(\omega)$ at $\sigma_c$. Moreover, the collapse of all data at $z > z_c$ implies that the value of $\sigma_c$ is independent of the packing fraction.

Now we see that solidlike states driven by a shear stress greater than $\sigma_c$ possess important features of marginally jammed solids, such as $z > z_c$ and jamminglike $D(\omega)$. It is thus plausible to strictly define them as shear jammed solids. The transition into shear jamming is driven by the shear stress, which resembles the jamming transition of frictionless and purely repulsive particles driven by the packing fraction, but now a critical-like point at $\phi_j$ is replaced by a range of packing fractions.

F. Jamming phase diagram

Our major findings lead to the jamming phase diagram for weakly attractive particles in the $\sigma$-$\phi$ plane at $T = 0$. Figure 5(b) is an example of the diagram for $\mu = 10^{-3}$.

Strictly defined shear jamming (SJ) is encircled by $\phi = \phi_{j,\mu}$, $\sigma_j(\phi)$, and $\sigma = \sigma_c$. Interestingly, $\sigma = \sigma_j$ intersects $\sigma_j(\phi)$ roughly at the rigidity percolation threshold $\phi_{rp}$. As mentioned earlier, solely from $S(q)$, it is hard to determine the crossover packing fraction to separate gel like states from jamminglike states. Now that SJ states only exist at $\phi > \phi_{rp}$, together with the evidence shown in Fig. 2(b), $\phi = \phi_{rp}$ is a plausible candidate of such a crossover.

Shear-driven solids lying below $\sigma = \sigma_c$ share some common features, e.g., existence of the low-frequency peak in $D(\omega)$, $z < z_c$, and attraction dominant, although they cover a wide range of packing fractions and exhibit progressive packing fraction evolution of the structure. States between $\phi_{rp}$ and $\phi_{j,\mu}$ are particularly interesting. They have similar structures to shear jammed states but resemble shear gel like states at $\phi < \phi_{rp}$ in mechanical and vibrational properties. We tentatively name them as intermediate states. The existence of intermediate states can only be found by the careful study of the shear stress dependence. It also warns us about the danger to identify various types of amorphous solids from structure [41] or vibrational properties [46] alone.

In order to verify that our major findings are not limited to specific potential and to two dimensions, in Figs. 5(c)–5(f), we show $z(\sigma)$ and the jamming phase diagram for two-dimensional Hertzian systems and for three-dimensional harmonic systems, respectively. Our major findings hold for all these systems.

G. Finite size effects

One may wonder if the existence of the SJ region discussed with the jamming phase diagram is a finite size effect. For purely repulsive particles, previous studies have shown that small size systems can jam below the critical packing fraction $\phi_j$ of the jamming transition. However, as an example shown in Fig. 1(a), the probability of finding jammed states in the absence of shear with $\mu = 10^{-3}$ is statistically zero when $\phi < 0.81$, while SJ states still survive below $\phi = 0.70$. Therefore, attraction and shear stress are both essential to the formation of SJ states, which is not simply due to finite size effects discussed above.

It has been shown that the yield stress of jammed solids of purely repulsive particles decreases with increasing system
increasing system size. For all system sizes studied, we find of shear-driven solids with attraction also decreases with a guide for the eye. (b) System size scaling of $\Delta \sigma = \sigma_\infty - \sigma_c$, i.e., the shear stress gap between the yield stress $\sigma_c$ and $\sigma_\infty$ in which SJ states exist, at $\phi = 0.75$ (circles) and 0.80 (squares). The solid lines are power-law scaling fits to the data: $\Delta \sigma - \Delta \sigma_\infty \sim N^{-1}$, where $\Delta \sigma_\infty = 2.26 \times 10^{-6}$ (6.90 $\times 10^{-6}$) for $\phi = 0.75$ (0.80).

size and approaches a limiting value in the large system size limit [23,24,43]. Figure 6(a) shows that the yield stress of shear-driven solids with attraction also decreases with increasing system size. For all system sizes studied, we find that the crossover shear stress $\sigma_c$, at which shear-driven solids are isostatic and have a flattened density of vibrational states is constant in packing fraction. With increasing system size, $\sigma_c$ decreases as well, but $\sigma_c(\phi)$ and $\sigma_c(\phi)$ always intersect approximately at $\phi = \phi_{\ast,\infty} \approx 0.689$, so the density interval for SJ states to exist seems almost insensitive to the change of system size. In Fig. 6(b), we show the system size dependence of $\Delta \sigma = \sigma_c - \sigma_\infty$ for SJ states at two different packing fractions. $\Delta \sigma$ at both packing fractions can be well fitted with the same power-law scaling: $\Delta \sigma = \Delta \sigma_\infty \sim N^{-1}$, where $\Delta \sigma_\infty$ is the extrapolation in the $N \rightarrow \infty$ limit. At both packing fractions, $\Delta \sigma_\infty > 0$. Therefore, our results suggest that SJ regime does not vanish in large systems.

IV. DISCUSSION AND CONCLUSIONS

In summary, in the presence of weak attraction, athermal solidlike states are explored by shear over a wide range of packing fractions below the jamming transition. Our careful study of the packing fraction and shear stress dependence reveals that the static structure of shear-driven solids is sensitive to the change of packing fraction, but not to shear stress. In contrast, the DOVS and force network evolve unexpectedly and progressively with shear stress. The strong shear stress dependence enables us to determine strictly defined shear jamming and construct a jamming phase diagram in the $\sigma$-$\phi$ plane.

As shear stress increases, the rigidity of shear-driven solids at $\phi < \phi_{\ast,\infty}$ increases, reflected in the decay of soft modes and increase of the coordination number and elastic moduli (not shown). In contrast, increasing shear stress slightly softens jammed solids well above $\phi_{\ast,\infty}$. This opposite behavior on both sides of $\phi_{\ast,\infty}$ is analogous to that of thermal systems: With increasing temperature, glasses are hardened below the jamminglike transition, while slightly softened above [47,48]. Therefore, we provide evidence supporting that the shear stress can have similar effects as the temperature on transitions between liquidlike and solidlike states and properties of amorphous solids [49,50], as proposed by the original jamming phase diagram [1].

Our work is relevant to experimental systems like granular materials and non-Brownian colloids. For colloidal systems with Brownian motion, how temperature affects shear-induced solidification is interesting to attack next. Both the thermal motion and shear can harden systems at $\phi < \phi_{\ast,\infty}$. It is quite interesting to figure out whether and how they may compete or help each other to induce unpredictable results in dynamics and phase behaviors.

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[31] http://www.caam.rice.edu/software/ARPACK.