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Growing length and time scales in a suspension of athermal particles

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We simulate a relaxation process of non-Brownian particles in a sheared viscous medium; the system is subject to the small shear strain and then undergoes relaxation. We estimate the exponents with which the relaxation time and the correlation length diverge as the density approaches the jamming density from below. In particular, the dynamic critical exponent is estimated as 4.6(2). It is also found that shear stress undergoes power-law decay at the jamming density, which is reminiscent of critical slowing down.

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In general (thermal or athermal) particulate systems at high density, the structural rearrangement is difficult due to the exclusion volume effect so that the structural relaxation time and the viscosity drastically increase. In particular, zero-temperature systems such as granular materials or emulsions acquire the elastic moduli above a certain density [1,2]. This rigidity transition, which is also referred to as the jamming transition, is accompanied by some power-law behaviors that are characteristic to critical phenomena, e.g., the growing correlation length in terms of spatially heterogeneous diffusion [3–7]. Indeed, such dynamical heterogeneities are also observed in thermal systems such as supercooled liquids and dense colloids [8–11]. This suggests the potential relation between the glass and jamming transitions, although still controversial [12–15].

However, the nature of the jamming transition itself is still not clear. Provided that jamming is a critical phenomenon, the critical exponents that describe the divergence of the length and time scales play an essential role in clarifying the mechanism of the dynamical heterogeneity. In addition, these exponents may classify the jamming transition into a universality class (if any). There are several attempts to estimate such exponents, in particular that for the correlation length; $\xi \sim |\phi - \phi_J|^{-\nu}$, where ϕ denotes the density and ϕ_J is the critical density. The finite-size scaling suggests that $\nu \simeq 0.7$ for both two- and three-dimensional systems [2]. This exponent is later confirmed by defining the correlation length in the dynamical heterogeneity [5–7,16,17]. Note that, however, these attempts on the dynamical heterogeneity exclusively involve two-dimensional systems. Recalling that the dimensionality plays an essential role in conventional critical phenomena, the correlation length in three-dimensional systems must be investigated.

Another important quantity that characterizes a critical phenomenon is the characteristic time. Wyart *et al.* [18] derived the characteristic frequency of jammed systems using the normal mode analysis to obtain $\omega_c \sim (\phi - \phi_J)^{\zeta}$, where $\zeta = 0.5$. However, note that the characteristic time in unjammed systems is still not estimated.

Along the line of thought, in this Rapid Communication, we investigate the behaviors of the relaxation time and the correlation length in a three-dimensional unjammed system at zero temperature. It is found that the relaxation time τ and the correlation length ξ increases obeying power laws with respect to the density; $\tau \sim (\phi_J - \phi)^{-3.3}$ and $\xi \sim (\phi_J - \phi)^{-0.7}$. These results lead to $z \simeq 4.6$, which coincides with that of a Lennard-Jones (LJ) glass [19].

We consider macroscopic particles in a viscous medium so that temperature does not play any role. Note the density is always smaller than the jamming density, as we focus unjammed systems here; $\phi < \phi_J \simeq 0.639$ [2]. We neglect the hydrodynamic and the electric interactions because we are interested in the nature of the jamming critical point. The particles are monodisperse, the diameter of which is denoted by d. Note that the particles are elastic so that the interaction between particles i and j is described by the linear repulsive force; i.e., $f_{ij} = k h_{ij} \mathbf{n}_{ij}$, where k denotes the elastic constant, $\mathbf{n}_{ij} = \mathbf{r}_{ij}/|\mathbf{r}_{ij}|$, and $h_{ij} = d - |\mathbf{r}_{ij}|$ denoting the overlap length. (Note that $h_{ij} = 0$ if $d < |\mathbf{r}_{ij}|$). Unless otherwise indicated, the system contains 6.4×10^4 particles.

A relaxation process is realized in such a way that an equilibrated system is perturbed at $t = 0$ and then relaxes. To prepare an initial equilibrated system under periodic boundary conditions, we adopt the conjugate gradient method, by which randomly distributed particles relax to a zero-energy state (no overlaps between particles). Then we perturb this equilibrated system by applying pure shear with respect to the (y,z) plane; i.e., each particle is instantaneously displaced by the following affine deformation,

$$y_i' = y_i + \epsilon z_i, \tag{1}$$

$$z_i' = z_i + \epsilon y_i, \tag{2}$$

where ϵ denotes the shear strain [20]. Accordingly, we adopt Lees-Edwards boundary conditions with respect to the y and z directions, in which the adjacent imaginary cells are displaced by ϵL. An ordinary periodic boundary condition is used along the x direction. Due to the shear strain, the overlap between particles appears so that the system acquires nonzero elastic energy. Then the system begins to relax and eventually reaches a new stable state of zero energy. In order to mimic the dynamics of macroscopic particles in a highly viscous medium, we adopt the overdamp dynamics; i.e., $\gamma \dot{\mathbf{r}}_i = \Sigma_j \mathbf{f}_{ij}$. Throughout this study, we adopt the units in which the mass, the diameter, and the mobility γ are unity. The elastic constant k is set to be γ^2/m. This procedure may be realizable in experiments using macroscopic particles in a viscous medium [21], where the shear strain is applied via the viscous medium and the gravity can be canceled by density matching.

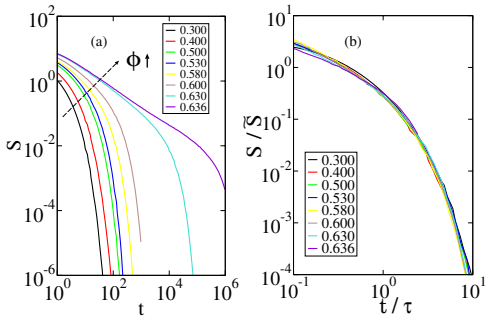

FIG. 1. (Color online) (a) The relaxation of shear stress at various densities (see the legends). Lower curves correspond to lower densities. The relaxation time grows as the density increases toward the critical density. Here the initial strain ϵ is 0.001. (b) Rescaled shear stress $S/\tilde{S}$ as a function of rescaled time, t/τ, where τ is the characteristic time that depends on the density.

First, we discuss the relaxation of macroscopic quantities, in particular the shear stress, which is defined via the virial [22]. Because other macroscopic quantities such as pressure or energy show essentially the same behavior, we focus the shear stress here. The relaxation of shear stress at each density is shown in Fig. 1(a), where the initial strain ϵ is 1.0×10^{-3}. We also test $\epsilon=0.01$ and $\epsilon=0.05$ to confirm that the result does not depends on the initial strain. Then the characteristic time is extracted from these relaxation behaviors by rescaling the time and the shear stress with τ and $\tilde{S}$, respectively. Then the relaxation profiles collapse onto a single curve as shown in Fig. 1(b). We find that this master curve can be described by

$$S(t) \sim S_0 t^{-\alpha} e^{-t/\tau}, \tag{3}$$

with $\alpha=0.55(5)$. The relaxation time τ drastically increases at higher densities as shown in Fig. 2(a). We confirm that the initial configuration does not significantly affect the relaxation behavior; less than $\pm10\%$ in the characteristic time. The result indicates that the relaxation time obeys a power law to diverge at the jamming density,

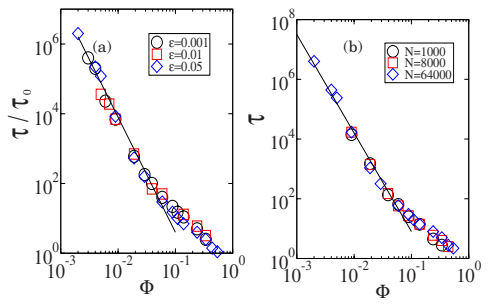

FIG. 2. (Color online) The relaxation time as a function of density. Solid line in each panel is proportional to $\tau\sim\Phi^{-3.3}$, where $\Phi=\phi_J-\phi$ and $\phi_J=0.639$. (a) The relaxation time normalized by τ_0: $\tau_0=2$ for $\epsilon=0.05$, $\tau_0=1$ otherwise. The power-law divergence and the exponent do not depend on the initial strain. (b) There is no system-size dependence of the relaxation time. The initial strain $\epsilon=0.05$.

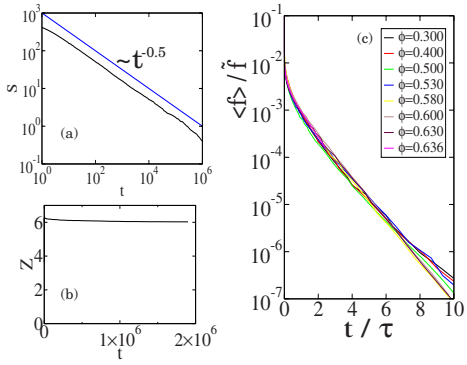

FIG. 3. (Color online) (a) The relaxation of shear stress for $\phi=0.637$ and $\epsilon=0.05$. The solid line is proportional to $t^{-0.5}$. (b) The relaxation of the average coordination number for $\phi=0.637$ and $\epsilon=0.05$. (c) The relaxation of the average magnitude of interparticle force, $\langle f\rangle$. Time is rescaled by $\tau(\phi)$ shown in Fig. 2(a). Note that $\langle f\rangle$ is also rescaled by $\tilde{f}\sim\tau^{0.3}$. The initial strain is 0.001.

$$\tau \sim \tau_0(\phi_J-\phi)^{-\zeta}, \tag{4}$$

where τ_0 denotes the time constant that does not depend on the density and the exponent is estimated as $\zeta=3.3(1)$.

However, this exponent is valid only in higher density region ($\phi\geq0.60$) and there is a crossover to a different power law in the lower density region, $\zeta=1.5(1)$. Note that this is not due to a finite-size effect as illustrated in Fig. 2(b), which shows the relaxation time for three different systems: $N=1000, 8000, 64\,000$. This is indeed due to the qualitative change in the particle dynamics, because the correlation function discussed below, Eq. (5), cannot detect any cooperative motion in the lower density region, $\phi<0.60$. However, as we focus on the critical nature of the jamming transition here, we do not further discuss this crossover.

From Eqs. (1) and (4), it is expected that the relaxation of shear stress is described by a simple power law at the jamming density, $S(t)\sim S_0 t^{-\alpha}$. Figure 3(a) shows the stress relaxation at $\phi=0.637$, which obeys a power law for a considerable duration (up to 6 orders of magnitude). We remark that this power-law relaxation, which is quite similar to critical slowing down, is consistent with a theory in which shear stress is an order parameter that undergoes marginal stability at the jamming transition point [23,24].

The slow relaxation is also apparent at the particle level. For example, we observe the average magnitude of interparticle forces defined as $\langle f\rangle=2\Sigma_{i>j}|\mathbf{f}_{ij}|/NZ$, where Z is the coordination number. Interestingly, Z is almost constant during the relaxation process as shown in Fig. 3(b). The relaxation profile of the average force magnitude is shown in Fig. 3(c), where we use the same relaxation time as that for the shear stress. We find that $\langle f\rangle\sim t^{-\beta}e^{-t/\tau}$ and the critical slowing down in the vicinity of the jamming density with the exponent $\beta\simeq0.3$. Furthermore, we observe that the energy and the pressure relax in the same manner; i.e., $P\propto t^{-\beta}e^{-t/\tau}$ and $E\propto t^{-2\beta}e^{-2t/\tau}$. This is indeed trivial because $P\sim2(NZ)^{-1}\langle f\rangle d/V$ and $E\sim2(NZ)^{-1}\langle f^2\rangle/kV$, where d and k denote the diameter and the stiffness of the particles, respectively. Note that f obeys an exponential distribution [25] so that $\langle f^2\rangle\sim\langle f\rangle^2$.

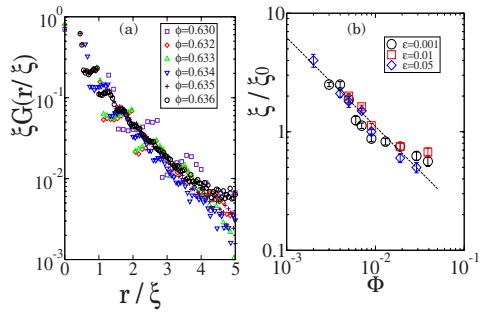

FIG. 4. (Color online) (a) The collapse of the correlation function defined by Eq. (5). Note that the length is normalized by the correlation length, ξ. (b) The correlation length as a function of $\Phi=\phi_J-\phi$, where $\phi_J=0.639$. The dashed line is proportional to $\Phi^{-0.75}$. Note that the correlation length is normalized by ξ_0, which depends on the initial strain ϵ. Here ξ_0 is set to be unity for $\epsilon=0.05$, while $\xi_0=0.8$ for $\epsilon=0.01$ and $\epsilon=0.001$.

We then investigate the spatial correlation of the particle motion during the relaxation process. To this end, we compare the initial and the final configurations to define the displacement vector of each particle; $\Delta\mathbf{x}_i=\mathbf{x}_i^{(1)}-\mathbf{x}_i^{(0)}-\mathbf{A}(\mathbf{x}_i^{(0)})$, where $\mathbf{x}_i^{(0)}$ and $\mathbf{x}_i^{(1)}$ are the initial and the final positions of particle i, and $\mathbf{A}(\mathbf{r})$ is the affine deformation vector at position $\mathbf{r}$, represented by Eqs. (1) and (2). Using the displacement vectors $\Delta\mathbf{x}_i$, we define the following correlation function:

$$G(r)=\frac{\sum_{i>j}\Delta\mathbf{x}_i\cdot\Delta\mathbf{x}_j\delta(r-|\mathbf{x}_i^{(0)}-\mathbf{x}_j^{(0)}|)}{\sum_{i>j}\delta(r-|\mathbf{x}_i^{(0)}-\mathbf{x}_j^{(0)}|)}, \tag{5}$$

which quantifies the extent of cooperative motion in dense particulate systems [6,9,26–29]. Figure 4(a) shows the collapse of the correlation function, where $\xi G(r/\xi)$ is plotted as a function of r/ξ. This indicates that ξ is the correlation length and the correlation function $G(r)$ is approximately exponential. As is shown in Fig. 4(b), this correlation length ξ increases as the density approaches the jamming density; $\xi\sim\xi_0(\phi_J-\phi)^{-\nu}$. The exponent ν is estimated as 0.73(5), which is indistinguishable from that of two-dimensional systems [3–7,16,17].

From these results we can estimate the dynamic critical exponent, $z=\zeta/\nu$. We replot the relaxation time as a function of the correlation length in Fig. 5, where we can estimate z as 4.6(2). This value is quite different from that obtained in two-dimensional air-fluidized beads, where $z\simeq 1.4$ [5]. At this point, we do not have any definite explanation on this difference: this may indicate the dimensionality dependence

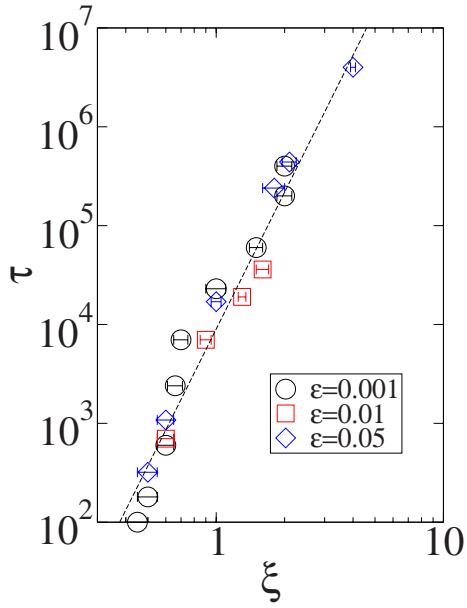

FIG. 5. (Color online) The relaxation time as a function of the correlation length. The dashed line is proportional to $\xi^{4.6}$.

of the dynamic critical exponent, or the structural relaxation process in athermal systems may depend on the specific agitating method by which energy is injected to the system. Nevertheless, it is noteworthy that almost the same dynamic critical exponent ($z\simeq 4.55\pm 0.2$) is found in a binary LJ supercooled liquid, where the correlation length and the characteristic time for the structural relaxation diverge toward zero temperature [19]. This result, together with a recent study [15], illustrates that the zero-temperature critical point potentially affects a class of supercooled liquids, although the equivalence of these exponents might be a mere coincidence.

To summarize, in a three-dimensional system at zero temperature, we show the power-law divergence of the relaxation time and the correlation length as the density approaches the jamming density from below; $\tau\sim\tau_0(\phi_J-\phi)^{-\zeta}$ and $\xi\sim\xi_0(\phi_J-\phi)^{-\nu}$, where $\zeta=3.3(1)$ and $\nu=0.73(5)$. The dynamic critical exponent is estimated as $z=4.6(2)$, which coincides with that of a supercooled liquid. We also observe the critical slowing down of the shear stress and the average force magnitude in the vicinity of the jamming density. Although the result is restricted to an unjammed system, we remark that the length and time scales in jammed systems and the effect of the dimensionality is currently investigated by using the present relaxation method.

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