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Effects of Cage-Breaking Events in Single-File Diffusion on Elongation Correlation

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Collective motion of caged particles is studied by calculating correlations of elongations (i.e. excess distances between two tagged particles) in a one-dimensional colloidal system, with the focus on the effect of overtaking events by which particles can hop out of the cage. It is shown analytically and verified numerically that the effect of overtaking is more prominent in shorter lengthscales, and also that the two-time elongation correlation exhibits ageing behavior due to overtaking.

KEYWORDS: single-file diffusion, overtaking, caged particles, collective motion, elongation correlation, Brownian dynamics, colloidal system, Rouse model, Lagrangian description, label variable

Various soft materials have properties between the liquid-like and solid-like consistencies, associated with collective dynamics of the constituents. As one of the simplest cases of such materials, one may mention a single elastic chain with thermal fluctuation, namely the Rouse model.[1,2] Indeed, the chain is not liquid nor solid in the usual sense: every monomer in the Rouse chain is inseparably bonded to its neighbors, yet its mean square displacement (MSD) can grow unlimitedly, in proportion to the square root of the elapsed time t.

The above-mentioned behavior of MSD $\propto \sqrt{t}$ is shared by one-dimensional (1D) systems of Brownian particles with repulsive interaction that disallows the particles to exchange their positions. This is known as the *single-file diffusion* (SFD).[3–5] In the ideal case in which the barrier height of the interaction potential, V_{max}, is infinitely large, every particle is eternally caged between its neighbors, and the longtime dynamics are equivalent to those of the Rouse model.[5,6] The growth of MSD is understood as a collective motion of particles comprising the cage, which is due to the dominance of long-wave fluctuations peculiar to low-dimensional systems and akin to the logarithmic behavior of the MSD in two-dimensional (2D) systems.[7–9]

More generally, collective motions in glassy dynamics of soft materials require four-point space-time correlations for their quantification.[10,11] Dynamical susceptibility χ_4 is one of such four-point correlations, developed for computational ease, though its behavior is rather difficult to interpret, as signals from different processes are mixed in it.[12–15] Some other types of space-time correlations are

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therefore needed. While the weakest point of χ_4 is that the effect of quasi-uniform cage drift appears as a decaying factor which obscures the growth of correlation length, it is known that this weakness can be overcome by space-time correlations based on the idea of particle tracking. In numerical analysis of glassy liquids, this idea has been implemented as bond breakage correlation,[13,16,17] which is completely frame-independent. Displacement correlation, which is also based on particle tracking, has the advantage of analytical tractability in some cases.[5,12,14,18,19]

Here we propose yet another space-time correlation, which at once inherits the strong points of the bond breakage correlation, being free from undesirable effect of drift, and allows analytical calculation in the same way as the displacement correlation. The idea is to target on correlations of *elongation*, analogous to the interparticle distance correlation previously studied by Lizana *et al.*[6] for ideal SFD on the basis of the elastic approximation (i.e. the Rouse model).

As an example to illustrate our calculation scheme of elongation correlation C_ε, going beyond the elastic approximation, here we take SFD with overtaking,[19–22] allowing the particles to hop out of the cage. Although there has been a number of studies on the crossover behavior of MSD (from $\sqrt{t}$ to t) in SFD with overtaking as a cage-breaking event, to the best of our knowledge, none of them have presented analytical calculation of space–time correlations to clarify the effect of overtaking on the collective motion. We establish a framework for analytical calculation of C_ε, extending the *label variable* method.[5,14,18,19,23] As a result, within a certain approximation justifiable in the limit of rare overtaking, C_ε is obtained as a sum of two parts: a contribution from the density fluctuation in the chain of the caged particles, and the effect of overtaking. At larger lengthscales the former predominates, while the latter has an impact on the shortscale behavior of C_ε.

The system is specified as follows: The position of the i-th particle, $X_i = X_i(t)$, is subject to the Langevin equation

$$m\ddot{X}_i = -\mu\dot{X}_i - \frac{\partial}{\partial X_i}\sum_{j<k} V(X_k - X_j) + \mu f_i(t), \tag{1}$$

where m is the mass of the particle, μ is the drag coefficient (a scalar constant), and $\mu f_i(t)$ is the random force characterized by the free-particle diffusivity $D = k_\mathrm{B}T/\mu$. The periodic boundary condition, $X_{j+N} = X_j + L$, implies the mean density $\rho_0 = N/L$. The interaction potential, determining the particle diameter σ, is specified as $V(\pm r) = V_\mathrm{max}\,(1 - r/\sigma)^2$ for $0 \le r \le \sigma$ and $V(r) = 0$ otherwise; V_max is large but finite, allowing neighboring particles to exchange their positions as a rare event.

On the basis of $\{X_i\}_{i=1,2,\dots,N}$, we define the *elongation* of the particle pair (i, j) relative to the initial configuration, as

$$\varepsilon_{i,j}(t) \overset{\mathrm{def}}{=} \frac{X_j(t) - X_i(t)}{X_j(0) - X_i(0)} - 1. \tag{2}$$

Its correlation, as a function of two time arguments s and t and the initial separation $\tilde{d}$ ($\neq 0$), is then introduced as

$$C_\varepsilon(\tilde{d}, t, s) \stackrel{\text{def}}{=} \tilde{d}^{\,2} \left\langle \varepsilon_{i,j}(t)\varepsilon_{i,j}(s) \right\rangle_{\tilde{d}} \quad (0 \leq s < t), \tag{3}$$

where $\langle \quad \rangle_{\tilde{d}}$ denotes conditional thermal averaging over the pairs (i, j) such that $X_j(0) - X_i(0) = \tilde{d}$.

For simplicity, here we limit the initial condition mainly to the equidistant configuration, $X_i(0) = X_0(0) + i\ell_0$ where $\ell_0 \stackrel{\text{def}}{=} L/N = \rho_0^{-1}$. In this case, Eq. (3) requires integer values of $\tilde{d}/\ell_0$, which we denote with Δ ($= \tilde{d}/\ell_0 \in \mathbb{Z}$), so that

$$\varepsilon_{i,i+\Delta}(t) = \frac{X_{i+\Delta}(t) - X_i(t)}{\ell_0\Delta} - 1. \tag{4}$$

As long as it is not confusing, we will write simply $C_\varepsilon(\Delta, t, s)$ instead of $C_\varepsilon(\ell_0\Delta, t, s)$. A variant of C_ε for a single time is also introduced analogously:

$$C^0_\varepsilon(\Delta, s) \stackrel{\text{def}}{=} \lim_{t\to s} C_\varepsilon(\Delta, t, s) = \frac{{\ell_0}^2\Delta^2}{N} \sum_i \left\langle [\varepsilon_{i,i+\Delta}(s)]^2 \right\rangle. \tag{5}$$

Theoretical approach to these statistical quantities is grounded on hydrodynamical field variables. The coarse-grained dynamics of $\{X_i\}_{i=1,2,\ldots,N}$ for timescales greater than m/μ are described by the Dean–Kawasaki equation,[24,25] with the fluctuating density field ρ and its flux Q defined as

$$\rho = \rho(x, t) = \sum_i \rho_i(x, t), \quad Q = Q(x, t) = \sum_i \rho_i(x, t)\dot{X}_i(t),$$

where $\rho_i(x, t) = \delta(x - X_i(t))$ is the single-body density.

Subsequently, we introduce the label variable ξ to incorporate the idea of particle tracking into the continuum description.[5,14,18,19,23] We define $\xi = \xi(x, t)$ as a solution to the equation $(\rho, Q) = (\partial_x\xi,\ -\partial_t\xi)$, so that ξ satisfies

$$\rho\,(\partial_t + u\,\partial_x)\,\xi(x, t) = 0, \tag{6}$$

with u such that $Q = \rho u$. The convective equation (6) implies that, if we define $\Xi_i(t) \stackrel{\text{def}}{=} \xi(X_i(t), t)$, its value is basically independent of t. In the absence of overtaking, $\Xi_i(t)$ is actually equivalent to the numbering of the particle. The constancy of $\Xi_i(t)$ is checked by calculating its t-derivative as[23]

$$\frac{\mathrm{d}\Xi_i(t)}{\mathrm{d}t} = \dot{X}_i(t)\left.\frac{\partial\xi}{\partial x}\right|_{x=X_i} + \left.\frac{\partial\xi}{\partial t}\right|_{x=X_i} = \left.\left(\rho\dot{X}_i - Q\right)\right|_{x=X_i}, \tag{7}$$

which should vanish unless some other particle, say the j-th one, overlaps the i-th particle. As a result of the overlap in exceptional cases, $\Xi_i(t)$ changes its value if the velocity difference $\dot{X}_i - \dot{X}_j \neq 0$ persists until the two particles exchange their positions: this is what we refer to as overtaking.

The overtaking event is thus formulated as a change in the values of $\Xi_i(t)$ and $\Xi_j(t)$, such that their values are exchanged: $\Xi_i(t_2) = \Xi_j(t_1)$ and $\Xi_j(t_2) = \Xi_i(t_1)$, where t_1 and t_2 denote times before and after the overtaking event. The absence of overtaking is expressed by $\mathrm{d}\Xi_i(t)/\mathrm{d}t = 0$, which could be interpreted as a kind of local conservation law for $\Xi_i(t)$ describing a topological constraint.

Apart from the overtaking event that allows a particle to hop out of the cage, the dynamics of each caged particle in the present system are governed by collective motion of the surrounding particles. This collective motion is described through density fluctuations; it is useful to express it with

$$\psi = \psi(\xi, t) \overset{\text{def}}{=} \ell_0^{-1} \frac{\partial x}{\partial \xi} - 1, \tag{8}$$

and introduce the Fourier representation, defined as

$$\psi(\xi, t) = \sum_k \check{\psi}(k, t) e^{-\mathrm{i}k\xi}, \quad \check{\psi}(k, t) = \int e^{\mathrm{i}k\xi} \psi(\xi, t) \frac{\mathrm{d}\xi}{N}, \tag{9}$$

where $k/(2\pi/N) \in \mathbb{Z}$. The field ψ is governed by a transformed version of the Dean–Kawasaki equation,[5,14,18,19,23] given as Eq. (2.12) in Ref. 14, whose linear approximation is found to be equivalent to the Rouse model[1,2,5] and the 1D Edwards–Wilkinson equation.[12,26] Let us thereby calculate the elongation correlation $C_\varepsilon(\Delta, t, s)$, defining[14,23]

$$C_\psi(k, t, s) \overset{\text{def}}{=} \frac{N}{L^2} \left\langle \check{\psi}(k, t) \check{\psi}(-k, s) \right\rangle. \tag{10}$$

We begin with expressing $x = x(\xi, t)$ as an indefinite integral of $\partial x/\partial \xi$. Using Eqs. (8) and (9), we find

$$x = x(\xi, t) = \ell_0 \xi + \ell_0 \sum_k e^{-\mathrm{i}k\xi} \frac{\check{\psi}(k, t)}{-\mathrm{i}k} + X_\mathrm{G}(t), \tag{11}$$

where $X_\mathrm{G}(t)$ is the center-of-mass fluctuation that vanishes for large systems.[5] Evaluation at $\xi = \Xi_j(t)$ then yields

$$X_j(t) = \ell_0 \Xi_j(t) + \ell_0 \sum_k e^{-\mathrm{i}k\Xi_j(t)} \frac{\check{\psi}(k, t)}{-\mathrm{i}k}. \tag{12}$$

From Eqs. (4) and (12), in principle, a formula to calculate C_ε from C_ψ can be derived. This derivation is carried out firstly in the case of the ideal SFD without overtaking, and secondly in the case in which overtaking is rare but not negligible.

In the first case, in which $\Xi_j(t)$ is frozen to its initial value $\Xi_j(0) = \Xi_j^0$ as overtaking is forbidden, Eq. (4) reads

$$\varepsilon_{j,j+\Delta}(t) = \frac{1}{\Delta} \sum_k e^{-\mathrm{i}k\Xi_j^0} \frac{e^{-\mathrm{i}k\Delta} - 1}{-\mathrm{i}k} \check{\psi}(k, t). \tag{13}$$

Then we multiply Eq. (13) with its duplicate in which (k, t) is replaced with $(-k', s)$, and expand the double summation. Taking into account that the terms with $k \neq k'$ vanish on the average, we arrive at the formula in the absence of overtaking:

$$C_\varepsilon(\Delta, t, s) = 2\ell_0^2 \sum_k \frac{1 - \cos k\Delta}{k^2} \left\langle \check{\psi}(k, t) \check{\psi}(-k, s) \right\rangle$$

$$\to \frac{L^4}{\pi N^2}\int_{-\infty}^{+\infty}\frac{1-\cos k\Delta}{k^2}C_\psi(k,t,s)\mathrm{d}k. \tag{14}$$

This formula (14) allows us to express C_ε more concretely, if a concrete expression for C_ψ is available. Here we use[5,26]

$$C_\psi(k,t,s) = \frac{S}{L^2}e^{-D_*^{\mathrm{c}}k^2(t-s)} + \frac{S_{\mathrm{init}}-S}{L^2}e^{-D_*^{\mathrm{c}}k^2(t+s)} \tag{15}$$

with $D_*^{\mathrm{c}} = \rho_0^2 D/S$, obtained from the linearized equation for ψ, with the non-equilibrium initial condition taken into account; S denotes the long-wave limiting value of the static structure factor in the equilibrium state, and S_{init} is the value corresponding to the initial condition ($S_{\mathrm{init}} = 0$ for the equidistant configuration). The integral in Eq. (14) is then evaluated, which yields

$$\frac{C_\varepsilon(\Delta,t,s)}{S\ell_0^2\Delta} = \varphi_\varepsilon\left(\frac{\Delta}{2\sqrt{D_*^{\mathrm{c}}(t-s)}}\right) - \varphi_\varepsilon\left(\frac{\Delta}{2\sqrt{D_*^{\mathrm{c}}(t+s)}}\right), \tag{16}$$

where the function $\varphi_\varepsilon(\,\cdot\,)$ is defined as

$$\varphi_\varepsilon(\theta) \overset{\mathrm{def}}{=} \operatorname{erf}\theta + \frac{-1+e^{-\theta^2}}{\sqrt{\pi}\,\theta} \simeq \begin{cases} \frac{\theta}{\sqrt{\pi}} - \frac{\theta^3}{6\sqrt{\pi}} + \cdots & (|\theta| \ll 1) \\ 1 - \frac{1}{\sqrt{\pi}\,\theta} & (\theta \to +\infty). \end{cases}$$

Notice the ageing effect in Eq. (16): if $t-s$ is fixed, still C_ε depends on s. In particular, for $t \to s$ we have

$$C_\varepsilon^0(\Delta,s) = S\ell_0^2\Delta\left[1-\varphi_\varepsilon(\theta^0)\right], \quad \theta^0 \overset{\mathrm{def}}{=} \frac{\Delta}{\sqrt{8D_*^{\mathrm{c}}s}}, \tag{17}$$

which is s-dependent (unless $s \gg \Delta^2/D_*^{\mathrm{c}}$). We also note that, if ageing is negligible ($s \gg \Delta^2/D_*^{\mathrm{c}}$ and $t-s \ll s$), Eq. (16) seems to be consistent with the result by Lizana *et al.*[6]

In Fig. 1, the values of C_ε^0 predicted by Eq. (17) are compared with those computed directly from Eq. (1). Except for the choice of $V(r)$ and the initial condition, the numerical calculation was performed in the same way as in Ref. 19, with finite inertia ($m/\mu : \sigma^2/D = 1 : 1$). The data in Fig. 1(a), plotted against θ^0, are seen to collapse onto a master curve given by Eq. (17), except for the systematic deviation at small values of θ^0. The case with the largest ρ_0 and the smallest $V_{\max}$, namely $(\rho_0, V_{\max}) = (0.5\sigma^{-1}, 10k_{\mathrm{B}}T)$, deviates most prominently. As this deviation is due to the omission of overtaking, now we need to proceed to the second case.

Let us discuss how the formula (14) is modified by overtaking, restarting from Eq. (12). For the sake of brevity, we define $\delta\Xi_j(t) \overset{\mathrm{def}}{=} \Xi_j(t) - \Xi_j(0) = \Xi_j(t) - \Xi_j^0$. On the assumption of rare overtaking, we regard $\delta\Xi_j(t)$ as a small perturbation, which allows linearizing Eq. (12) in $(\check{\psi}, \delta\Xi_j)$ as

$$X_j(t) \simeq \ell_0\Xi_j^0 + \ell_0\delta\Xi_j(t) + \ell_0\sum_k e^{-\mathrm{i}k\Xi_j^0}\frac{\check{\psi}(k,t)}{-\mathrm{i}k}. \tag{18}$$

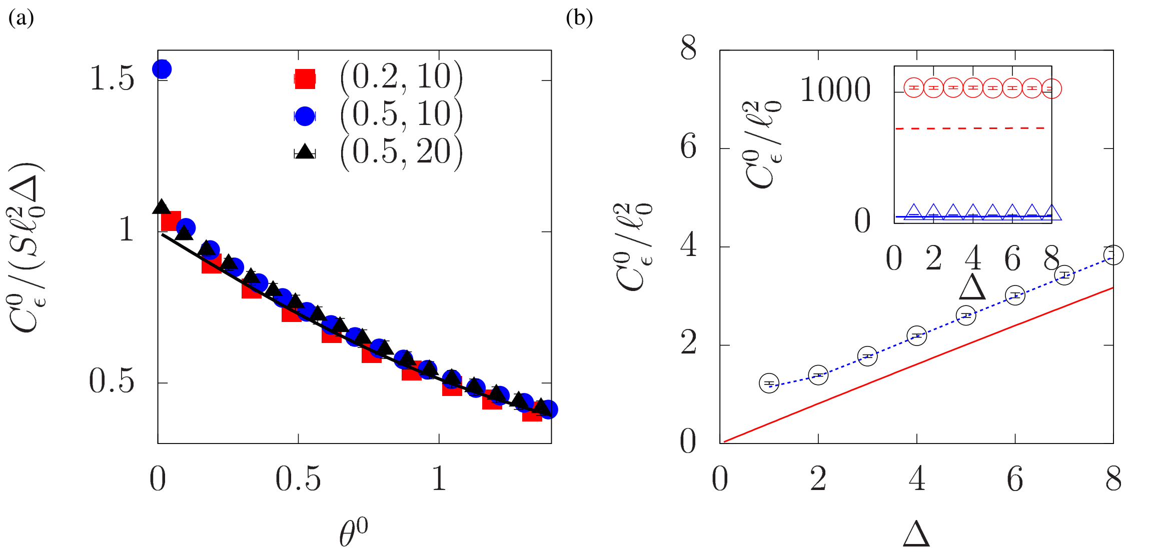

Fig. 1. (Color online) Numerical values of C^0_ε. (a) A plot against $\theta^0 = \Delta/\sqrt{8D^c_* s}$, rescaled with $S\ell_0^2\Delta$. Three cases are included: $(\rho_0\sigma, \beta V_{\max}) = (0.2, 10)$, $(0.5, 10)$ and $(0.5, 20)$. In each case, data are computed with $N = 500$, recorded at $s = 1000\,\sigma^2/D$, and averaged over 480 runs for each plotted point. The solid line represents the master curve predicted by Eq. (17). (b) A replot of $C^0_\varepsilon(\Delta, s)/\ell_0^2$ versus Δ, for $(\rho_0\sigma, \beta V_{\max}) = (0.5, 10)$ and $s = 4000\,\sigma^2/D$, shown with circles (∘). The lines represent theoretical predictions: the solid line represents Eq. (17), while the dotted line is given by Eq. (25) with H^0 taken into account, where $\nu_\alpha = 3.15 \times 10^{-5} D/\sigma^2$ according to Eq. (22). Inset: an analogous plot with $\beta V_{\max} = 2$ (broken line for the theory and circles for numerical values) and $\beta V_{\max} = 5$ (solid line and triangles).

Then, following the same line of argument as in the derivation of Eq. (14), we find

$$C_\varepsilon(\Delta, t, s) = 2\ell_0^2 \sum_k \frac{1 - \cos k\Delta}{k^2} \left\langle \check{\psi}(k, t)\check{\psi}(-k, s) \right\rangle + \ell_0^2 H(\Delta, t, s) \tag{19}$$

where $H(\Delta, t, s) \overset{\text{def}}{=} \left\langle \left[\delta\Xi_j(t) - \delta\Xi_i(t)\right]\left[\delta\Xi_j(s) - \delta\Xi_i(s)\right] \right\rangle_{j-i=\Delta}$.

The first term on the right-hand side of Eq. (19) reproduces Eq. (16), while the second term needs to be evaluated separately. To be consistent with the treatment of ψ based on the Dean–Kawasaki equation, the overtaking process should be treated on the basis of Dean's equation[24] for $\rho_i(x, t)$. However, within the approximation of the present analysis, a phenomenological modeling will suffice.

We model the overtaking as a random process in which a particle is exchanged with its neighbor at the frequency ν_α, such that $\left\langle [\delta\Xi_j(t) - \delta\Xi_j(s)]^2 \right\rangle = 2\nu_\alpha(t - s)$. Assuming that distinct exchanges are uncorrelated, we have

$$H(\Delta, t, s) = H(\Delta, s, s) \overset{\text{def}}{=} H^0(\Delta, s) \quad (0 \le s < t). \tag{20}$$

This is further evaluated by calculating the pair distribution function for $(\delta\Xi_i(s), \delta\Xi_{i+\Delta}(s))$, as

$$H^0(\Delta, s) = \mathfrak{s} + \mathfrak{s}\, e^{-\mathfrak{s}} \left[I_{\Delta-1}(\mathfrak{s}) + I_\Delta(\mathfrak{s})\right] + (-2\Delta + 1)e^{-\mathfrak{s}} \sum_{n=\Delta}^{\infty} I_n(\mathfrak{s}), \tag{21}$$

where $\mathfrak{s} = 4\nu_\alpha s$ and I_n denotes the modified Bessel function of the first kind. In particular, for $\Delta \gg 1$, Eq. (21) can be approximated by the first term alone, i.e. $H^0(\Delta, s) \simeq 4\nu_\alpha s$.

The overtaking frequency ν_α is given by an Arrenius-like expression, with a prefactor that depends on both the barrier hight $V_{\max}$ and the mean density ρ_0. In the present system, numerical data of ν_α can be fitted by

$$\nu_\alpha = D\left(a_0\frac{\rho_0}{\sigma} + a_1\rho_0^2\beta V_{\max}\right)e^{-\beta V_{\max}}, \quad \beta = \frac{1}{k_{\mathrm{B}}T}, \tag{22}$$

with $a_0 \approx 1/2$ and $a_1 \approx 1/6$.

The prediction by Eq. (19) is summarized as follows: Using the similarity variables

$$\theta \stackrel{\text{def}}{=} \frac{\Delta}{2\sqrt{D_*^{\mathrm{c}}(t-s)}}, \quad \theta' \stackrel{\text{def}}{=} \frac{\Delta}{2\sqrt{D_*^{\mathrm{c}}(t+s)}} \tag{23}$$

suggested by Eq. (16), we find Eq. (19) to predict

$$C_\varepsilon(\Delta, t, s) = S\,\ell_0^2\Delta\,[\varphi_\varepsilon(\theta) - \varphi_\varepsilon(\theta')] + \ell_0^2 H^0(\Delta, s), \tag{24}$$

with the hopping term estimated by Eqs. (21) and (22). In the limit of $t \to s$, we also have

$$C_\varepsilon^0(\Delta, s) = S\,\ell_0^2\Delta\left[1 - \varphi_\varepsilon(\theta^0)\right] + \ell_0^2 H^0(\Delta, s) \tag{25}$$

in place of Eq. (17). Let us test these predictions.

The prominent deviation from Eq. (17), seen in Fig. 1(a) for $(\rho_0, V_{\max}) = (0.5\sigma^{-1}, 10k_{\mathrm{B}}T)$, is clarified on the basis of Eq. (25). The replot in Fig. 1(b) exhibits a nearly uniform deviation from Eq. (17), attributable to the last term in Eq. (25) and consistent with Eq. (21). Note, however, that quantitative agreement is lost if $V_{\max}$ is so low that frequent overtaking events invalidate the present theory; see the Inset of Fig. 1(b).

The t-dependence of $C_\varepsilon = C_\varepsilon(\Delta, t, s)$ predicted by Eq. (24) is verified in Fig. 2, where C_ε is plotted against $t - s$. We have chosen a large value of s, so that $\varphi_\varepsilon(\theta')$ in Eq. (24) is negligible. In the case of $V_{\max} = 20k_{\mathrm{B}}T$, the barrier is so high that the hopping term $\ell_0^2 H^0(\Delta, s)$ is also negligible; this means that C_ε is given by $\varphi_\varepsilon(\theta)$ alone, as is shown by the lower solid line in Fig. 2, and C_ε decays away for $t - s \to +\infty$. Contrastively, if the barrier is lower, C_ε remains finite for $t - s \to +\infty$, as the hopping term contributes to it. We have evaluated $\lim_{t-s\to\infty} C_\varepsilon(\Delta, t, s)$ by means of fitting, as is exemplified by the dashed line in Fig. 2. The residual values thus obtained, denoted with C_ε^∞ and normalized with ℓ_0^2, are plotted against $4\nu_\alpha s$ in the inset of Fig. 2, with ν_α given by Eq. (22). The result seems to be reasonably close to the theoretical prediction, $C_\varepsilon^\infty/\ell_0^2 \simeq 4\nu_\alpha s$.

Thus we have presented a scheme to calculate C_ε in SFD with overtaking, by expressing the motion of particles in terms of $\check{\psi}(k, t)$ and $\delta\Xi_j(t)$. The field $\check{\psi}(k, t)$ represents fluctuation of density waves, while $\delta\Xi_j(t)$ is a locally conserved quantity serving as an indicator of overtaking. Within the

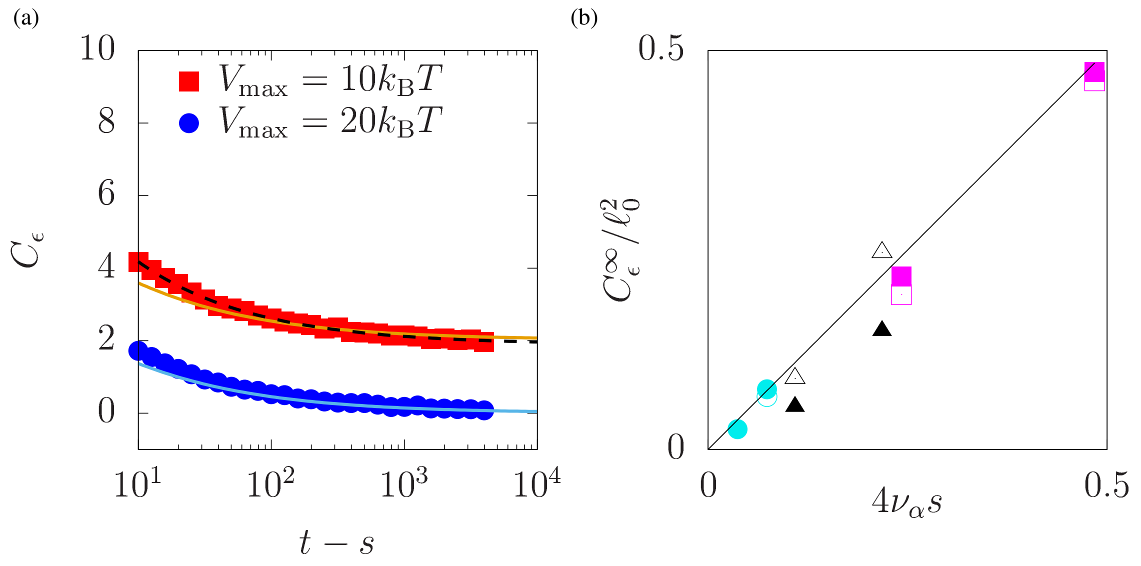

Fig. 2. (Color online) (a) Decay of $C_\varepsilon(\Delta, t, s)$ with regard to t. The two cases of $V_{\max} = 10k_BT$ and $V_{\max} = 20k_BT$ (with $\rho_0 = 0.5\sigma^{-1}$ in common) are compared by plotting C_ε against $t - s$, with $s = 4000\,\sigma^2/D$ and $\Delta = 3$ fixed; 160 runs are averaged in each case. The solid lines represent theoretical curves predicted by Eq. (24), and the dashed line results from fitting with $A + B(t-s)^{-1/2}$. (b) Numerical values of $C_\varepsilon^\infty = \lim_{t-s\to\infty} C_\varepsilon(\Delta, t, s)$, nondimensionalized with ℓ_0^2 and plotted against $4\nu_\alpha s$, i.e. the leading term in Eq. (21). The squares, circles and triangles denote $(\rho_0\sigma, \beta V_{\max}) = (0.5, 10)$, $(0.5, 12)$ and $(0.3, 10)$, respectively; the filled symbols represent results for $\Delta = 3$, and the open ones for $\Delta = 5$.

linear approximation, C_ε is obtained in Eq. (24) as a sum of two contributions from $\check{\psi}$ and $\delta\Xi_j$. The effect of overtaking is prominent at shorter lengthscales, but it has a relatively small impact on the long-range correlation (Fig. 1). This is naturally understood, on one hand, by considering that the overtaking process in SFD is a short-scale event involving only two neighboring particles explicitly. This interpretation suggests, on the other hand, that it will be quite intriguing to extend the present framework to systems in which cage-breaking events involve many particles, as the result will provide information about the space-time scales of such events.

The hopping term $\ell_0^2 H^0(\Delta, s)$ in Eqs. (24) and (25) depends on s and grows unlimitedly. This means that C_ε never equilibrates: C_ε is subject to an extra ageing effect due to overtaking, in addition to the effect of $S_{\text{init}} \neq S$ on C_ψ in Eq. (15). Besides, the temporal behavior of H^0 and C_ψ in the present system are quite similar to that of the correlations of rotational and dilatational modes of deformation in 2D colloidal liquids.[5,18] On this analogy, we expect that more insight may be given by profound studies of overtaking: for example, the ρ_0-dependence of ν_α in Eq. (22) may be clarified by ideas in Ref. 22 and suggest an extension to 2D colloidal glasses.

In a wider context of glassy dynamics, cage-breaking events can be conceived as transition between configurations corresponding to local basins of the energy landscape, often termed as inherent structures.[27] The overtaking event in SFD is among the simplest examples of such transition. As is shown in Fig. 2, the effect of this transition between inherent structures remains in C_ε for $t - s \to \infty$. In other words, the "natural distance" between the two tagged particles has changed from its initial value. This is reminiscent of the theory of elastoplasticity in terms of natural metric,[28] which may help to clarify the Nakahara–Matsuo memory effect in pastes[29] as a manifestation of stress anisotropy induced by shaking.[30] An extension of the present work in the direction of these studies on granular pastes[28–30] might be possible, if the change in C_ε is related to the stress field in some way analogous to nonlinear interaction between $\check{\psi}$ and $\delta\Xi_j$.

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