Dynamical phase transitions in one-dimensional hard-particle systems

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We analyze a one-dimensional model of hard particles, within ensembles of trajectories that are conditioned (or biased) to atypical values of the time-averaged dynamical activity. We analyze two phenomena that are associated with these large deviations of the activity: phase separation (at low activity) and the formation of hyperuniform states (at high activity). We consider a version of the model which operates at constant volume, and a version at constant pressure. In these nonequilibrium systems, differences arise between the two ensembles, because of the extra freedom available to the constant-pressure system, which can change its total density. We discuss the relationships between different ensembles, mechanical equilibrium, and the probability cost of rare density fluctuations.

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I. INTRODUCTION

Out-of-equilibrium systems differ from their equilibrium counterparts in many ways. For example, long-ranged correlations are generic in nonequilibrium steady states [1,2]; unusual phase transitions can take place [3]; and there may be significant differences in behavior for the same system in different ensembles (for example, canonical and grand canonical [4,5]). Recently, there has been considerable interest in large-deviation phenomena [6], based on ensembles of trajectories that are conditioned on atypical values of time-averaged observables [7–12]. In glassy systems, ensembles conditioned on the dynamical activity quite generically support phase transitions between active and inactive states [9,13]. In other systems, such biases can result in phase separation [14], and other kinds of phase transition [7].

Here, we consider large deviations of the activity in a model of hard particles that diffuse in one dimension. Considering this system in a constant-volume ensemble, we showed previously [15] that a bias to lower than average activity leads to phase-separated states, in which the density becomes macroscopically inhomogeneous. Also, on biasing to higher than average activity, we found that the system enters a hyperuniform state, in which large-scale density fluctuations are strongly suppressed [16]. On large length and time scales, the system can be described by fluctuating hydrodynamics and the macroscopic fluctuation theory [17,18], and both phase separation [14] and hyperuniformity [15] can be predicted from these theories.

In this work, we extend these results in three ways. First, we analyze the phase-separated state in more detail, including a finite-size scaling analysis, an analysis of the balance of forces at the interface between the dense and dilute phases, and the stability of the phase-separated state. Second, we consider the same hard-particle system in a constant-pressure ensemble. That is, we allow the system size to fluctuate, coupling it to a barostat that enforces a given mechanical pressure. For the system at equilibrium, the implementation of the barostat is standard [19], and is chosen so that the constant-pressure system resembles a typical subsystem of a very large constant-volume system. However, on biasing the system to nontypical dynamical activity, the constant-pressure system no longer mirrors the behavior of a subsystem of the constant-volume one: for a bias to low activity, the constant-volume system exhibits phase coexistence between dense and dilute phases, but the constant-pressure one has only a dense phase. For high activity, hyperuniformity is less apparent in the constant-pressure system, which tends instead to reduce its total density. We discuss how equivalence between constant-pressure and constant-volume ensembles should be interpreted in systems exhibiting large deviations.

Third, we analyze the mechanical pressure within these systems, as a function of the bias to low (or high) activity. The pressure is defined by analogy with the virial pressure in equilibrium systems, by considering the force exerted on one region of the system by other neighboring regions. In the phase-separated state, this pressure is not constant throughout the system, in stark contrast to the situation in equilibrium systems at phase coexistence. Describing the system in terms of a Langevin equation, we show how pressure gradients are balanced by noise forces that acquire finite average values, due to the low-activity bias on the system. Similar phase coexistence phenomena appear in active matter systems: we argue that particles’ swimming forces in those systems act similarly to biased noises, and can balance pressure gradients [20–23].

Taken together, our results highlight the broad range of phenomena that can occur in conditioned (or biased) ensembles of trajectories, even in simple models. They show that equilibrium ideas of pressure and ensemble equivalence can sometimes be applied in these contexts, but that these applications may be subtle and require careful justification. In the following, Sec. II describes the models and methods that we will use, Sec. III shows results for constant-volume systems, while Sec. IV includes results at constant pressure. In Sec. V, we discuss force balance and mechanical equilibrium in these systems. We draw our conclusions in Sec. VI.

II. MODEL AND METHODS

A. Model

1. Constant volume

We consider $N$ hard particles undergoing Brownian motion in one dimension, with periodic boundaries. Particle motion is
described by the overdamped Langevin equation:
\[
\dot{x}_i = -\frac{D_p}{k_B T} \nabla_i U + \sqrt{2D_p} \eta_i(t),
\]
where \(x_i\) is the position of particle \(i\), \(U\) is the potential energy of the system, \(D_p\) is the diffusion coefficient for particle motion, and \(\nabla_i = (\partial/\partial x_i)\). The notation \(\dot{x} \equiv (\partial x/\partial t)\) and we use Itô’s convention for stochastic differential equations throughout [24]. The random noise \(\eta_i\) has zero mean and is uncorrelated in time and space:
\[
\langle \eta_i(t)\eta_j(t') \rangle = \delta(t - t')\delta_{ij}.
\]
One may easily show [24] that the steady state of this system is a Boltzmann distribution \(p(x) \propto \exp[-U(x)/k_BT]\).

We consider the case where the energy \(U\) is given by a pair potential, \(U = \frac{1}{2} \sum_{i<j} v(x_i - x_j)\). Specifically, we consider hard particles of size \(l_0\),
\[
v(x) = \begin{cases} 
\infty, & \text{if } |x| \leq l_0, \\
0, & \text{otherwise}.
\end{cases}
\]
In this case, particles cannot interpenetrate or otherwise move past one another, leading to single-file diffusion [25]. [For the derivatives in (1) to make sense, one should regularize the potential by smoothing its discontinuities, so that the hard-particle case may be obtained by taking a suitable limit. In practice, we use a Monte Carlo scheme to solve the equation, which avoids the requirement for any explicit regularization.]

In one dimension, the equation of state for this system is simply that of an ideal gas with excluded volume:
\[
P(L - Nl_0) = Nk_BT,
\]
confirming that the steady state of the system has an (average) virial pressure equal to \(P\), as required. For large systems, the system size is almost always close to its mean value \(\bar{L} = \langle L \rangle\), with small fluctuations of size \(O(\sqrt{N})\). Further, if the system size \(L\) fluctuates to a value \(\bar{L} + \delta L\) that is slightly larger than its average value then the virial pressure of the system will be \(P \approx P - \delta L/\bar{L}k_BT\), where \(k_T = (-1/\bar{L})\delta L/\partial P\) is the isothermal compressibility. Inserting this relation into (6) and taking the average over the stochastic dynamics, one arrives at a deterministic equation for the relaxation of this system-size fluctuation \(\delta L = \frac{1}{\tau_L} \delta L\). Solving this equation, one sees that fluctuations in the system size \(L\) relax on a time scale \(\tau_{LL} = (\bar{L}k_Tk_BT/D_L)\).

To fix the value of \(D_L\), we note that for a large constant-volume system, density fluctuations on a length scale \(R\) should relax on a time scale of order \(\tau_{LL}(R) = R^2/(2\pi \tau_L^2 D_L)\), where \(D_L\) is a collective diffusion constant. We assume that \(D_L\) is similar to the bare particle diffusion constant \(D_p\) (this assumption can be justified within a fluctuating hydrodynamic or macroscopic fluctuation theory approach [17,27]). We then match time scales between constant-volume and constant pressure systems by setting \(\tau_{LL} = \tau_L(\bar{L})\), which yields
\[
D_L \approx \frac{(2\pi)^2 D_p k_T k_B T}{\bar{L}}.
\]

The numerical prefactor in this equation is not crucial for this work but the scaling of \(D_L\) with system size \(\bar{L}\) will be important in what follows.
3. Monte Carlo dynamics

In our numerical simulations of this model, we use a Monte Carlo dynamical scheme. Details are given in Appendix A. The scheme includes a time step \( \tau_0 \) as an explicit parameter, and the maximal displacement of a particle in a single MC move is \( S = \sqrt{6D_0\tau_0} \). In the limit of small \( \tau_0 \), the MC method is equivalent to solving the Langevin equations (1) and (6).

B. Large deviations

As anticipated in the Introduction, we will be concerned here with ensembles of trajectories which are biased to nontypical values of an “activity” parameter \( K \). The methodology that we use is now well established (see for example [7,9,10,13]): our specific approach is described in this section and follows closely that of [9]. A trajectory of the system is a realization of the system developing through time. A trajectory has a time duration \( \tau_{obs} \), which is composed of \( M \) smaller and consecutive “slices” of time, each of length \( \Delta t \), such that \( \tau_{obs} = M \Delta t \).

To define the activity \( K \), we measure the squared displacements of all particles during each slice and sum over the \( M \) slices in a trajectory:

\[
K[x(t)] = \sum_{j=1}^{M} \sum_{i=1}^{N} [x_i(j\Delta t) - x_i((j-1)\Delta t) - \Delta \bar{x}_j]^2,
\]

(9)

where the notation \([x(t)]\) indicates a functional dependence on all particle positions throughout a trajectory. The quantity \( \Delta \bar{x}_j = (1/N) \sum_i [x_i(j\Delta t) - x_i((j-1)\Delta t)] \) is the change of the center of mass position during the \( j \)th slice: for large systems this term has a negligible effect on \( K \) but subtracting it in this way helps to minimize finite-size effects in simulations. It is often useful to normalize the activity as

\[
k = \frac{K}{L\tau_{obs}}.
\]

(10)

(For the constant pressure systems, we replace \( L \) by \( \bar{L} \) in this equation.) We take the time interval \( \Delta t = \tau_B \), so that \( K \) depends on movement of particles on length scales comparable with their size, consistent with [9]. Figure 1 shows the dependence of the activity \( k \) on the packing fraction \( \phi \) and the maximum MC step \( S \) in the constant-volume ensemble. The activity is linear in \( \phi \) for small packing fractions, because \( k \) is proportional to the number of particles per unit volume. For larger \( \phi \), particles start to obstruct each other and the activity falls.

![Graph showing activity as a function of packing fraction and maximum MC step size](image)

**FIG. 1.** (Color online) Equilibrium activity \( k \) as a function of the packing fraction \( \phi \) and the maximum MC step \( S \) in the constant-volume ensemble. The activity is linear in \( \phi \) for small packing fractions, because \( k \) is proportional to the number of particles per unit volume. For larger \( \phi \), particles start to obstruct each other and the activity falls.

The large deviation formalism uses a biasing field \( s \) to investigate trajectories where \( \tau_{obs} \) is large but the activity \( k \) deviates significantly from its typical value. To this end, we define a probability distribution over the trajectories of the system:

\[
P_s[x(t)] = \frac{P_0[x(t)]e^{-\Delta K[x(t)]}}{Z(s,\tau_{obs})},
\]

(11)

where \( Z(s,\tau_{obs}) = e^{-\Delta K_{eq}} \) is a dynamical partition function (the average is evaluated at equilibrium). By analogy with equilibrium thermodynamics, we interpret \( \psi(s,\tau_{obs}) = -(K_{obs})^{-1\infty}Z(s,\tau_{obs}) \) as a dynamical free energy (or free energy density). For large \( L \) and \( \tau_{obs} \), the free energy may develop a singular dependence on \( s \), which signals the presence of phase transitions.

We use a notation \((\cdot)_{s}\) to indicate averages with respect to the distribution (11). The free energy \( \psi(s) \) is a scaled cumulant generating function, so it is easily verified that the mean activity \( k(s,\tau_{obs}) \equiv (L\tau_{obs})^{-1}(K)_{s} \) is obtained by taking a derivative: \( k(s) = \partial\psi/\partial s \), while the susceptibility \( \chi(s,\tau_{obs}) \equiv (L\tau_{obs})^{-1\infty}(\langle K \rangle_{s} - \langle K \rangle)_{s} \) is equal to \(-\partial\psi/\partial s\). For large \( \tau_{obs} \), taking averages with respect to the distribution (11) is equivalent to studying trajectories of that system that are conditioned to (nontypical) values of the activity \( k \) [11,12]. The equivalence is similar to that between microcanonical and canonical ensembles in equilibrium statistical mechanics, and means that the main results shown here could also be obtained by considering trajectories conditioned on \( K \). The approach using the bias \( s \) is more convenient both computationally and analytically; see for example [2,7–9].

C. Transition path sampling

To sample the biased ensemble of trajectories given in (11), we use transition path sampling [28,29], following the implementation of [9,10]. An initial trajectory is generated from an equilibrium initial configuration by simulating the dynamics of a system for a duration \( \tau_{obs} \). A new trajectory is created by copying a randomly selected portion of the first trajectory to either the start or end of the new trajectory. The rest of the new trajectory is then generated according to the relevant Langevin equations. The new trajectory is compared to the first and replaces it with probability

\[
P_{accept} = \min\{1,e^{-\Delta K}\},
\]

(12)

where \( \Delta K = K_{new} - K_{old} \). Generation then continues using the most recently accepted trajectory as the parent. Using this method means that, after many iterations, the algorithm samples trajectories according to the distribution defined in (11).

III. RESULTS: CONSTANT-VOLUME SYSTEM

In this section we present results for biased ensembles of the hard-particle system in the \( NVT \) (constant volume) ensemble. All results are for the case \( \phi = 0.88 \), as in [15], which is a relatively high density: it gives a mean free space per particle of \( 0.136\tau_0 \) and a typical time between collisions of \( 0.018\tau_0 \).
This is a diffusive system and is described by the theory of fluctuating hydrodynamics (or macroscopic fluctuation theory) [17,18]. Based on this theory, we expect qualitative aspects of the system’s behavior to be independent of $\phi$ [15].

### A. Phase separation for $s > 0$

We first consider the effect of a bias $s > 0$, which leads to a phase transition in this system [15]. Phase transitions are signalled by singularities in the free energy $\psi(s)$, which appear only in the limit when both the observation time $t_{obs}$ and the system size $N$ are very large. Figure 2 shows the average activity $k(s)$ for different system sizes and observation times. In particular, Fig. 2(a) shows the effect of increasing $t_{obs}$ at fixed system size $N = 100$, while Fig. 2(b) shows dependence on system size $N$, all obtained for large $t_{obs} = 20\tau_B$. Similar results were shown in [15]: here we analyze this behavior in more detail.

As in glass-forming systems [9,13], biasing to low dynamical activity yields a crossover at some $s = s^*$, from active to inactive dynamics. The value of $s^*$ is positive and depends on both $N$ and $t_{obs}$. The properties of the crossover can be obtained by a finite-size scaling analysis, which involves a joint limit of large system size $N$ and large observation time $t_{obs}$ [9,14,30,31]. In this limit, the crossover becomes a sharp discontinuity in $k(s)$. To rationalize this behavior, note that for $s \to \infty$, the system must arrive at the state with minimal propensity for activity [15], which is the fully phase-separated state, where the particles in the dense cluster are all touching each other. It is therefore clear that phase separation must occur at some field $s^*$. In fact, this phase transition can be predicted and analyzed in detail in the framework of fluctuating hydrodynamics [1,17,32], which predicts that the system will phase separate whenever $(\partial^2/\partial\rho^2)(K)_{eq} < 0$, and that $s^* \sim N^{-2}$ tends to zero as the system gets large [14,15,27].

Figure 3 shows a finite-size scaling analysis of the transition. For fixed $t_{obs} = 20\tau_B$ and increasing $N$, we find $s^* \sim N^{-1}$, as indicated by the collapse of $k(s)$ and the scaling of the peak in the susceptibility $\chi$. It seems that the theoretical prediction that $s^* \sim N^{-2}$ [14,27] may be observable only for larger-$N$ and/or larger $t_{obs}$: we discuss this possibility in Sec. III C.

### B. Structure of the phase-separated state

Figure 4 shows trajectories from biased ensembles, both at equilibrium and for an inactive state. At $s = 0$ the system is an equilibrium fluid of hard particles. In the inactive phase [$s > s^*$, Fig. 4(b)] the system phase separates throughout the whole trajectory: one observes a significant region that is devoid of particles, with the remainder of the system increasing its local density to incorporate the empty region. At the beginning and end of the trajectory, the empty region shrinks slightly: this behavior is also expected, since biases on dynamical activity have their strongest effects in the bulk of the trajectory, with slightly weaker influence near the initial and final times [30].

To probe the structure of these systems it is convenient to make a change of coordinates. The system is one dimensional and the particles are hard, so the ordering of the particle coordinates is fixed: there is no “overtaking.” We number particles so that their coordinates are in an increasing sequence, and define new coordinates $X_j = x_j - j l_0$ which are also ordered.
in the same way. The $X_j$ are coordinates of point particles in a system of size $L' = L - N l_0$. At equilibrium, the positions $X_j$ are uncorrelated—they represent positions of ideal gas particles. For example, if we define a Fourier-transformed density $\delta \rho_q = \sum_j e^{-i q X_j}$ and calculate the structure factor

$$S(q) = \frac{1}{L} \langle \delta \rho_q \delta \rho_{-q} \rangle$$

then we find $S(q) = N/L' = \frac{\phi}{\phi(1 - \phi)}$, independent of $q$.

Now define $d_i$ as the separation between particle $i$ and its right neighbor,

$$d_i = x_{i+1} - x_i.$$  \hspace{1cm} (14)

In order to investigate the one-body density profile associated with the phase-separated state illustrated in Fig. 4(b), it is necessary to fix an origin. We accomplish this by finding the largest “gap” $d_i$ in any configuration. We choose a random point within that gap, and we place the origin the maximal possible distance, $L'/2$, from that point. Thus the origin almost certainly lies within the dense phase. The density of point particles is then $\rho(X) = \sum_j \delta(X - X_j)$, where $X_j$ is now measured with respect to this new origin. We average $\rho(X)$ to obtain the one-body densities shown in Fig. 5(a). At equilibrium, the density profile is uniform, as expected (up to weak boundary effects that arise because the origin was constrained to lie far from the largest gap).

The distribution of separations, $P(d)$, was also recorded for systems at $s = 0$ and $s > s^*$. This is shown in Fig. 5(b). At equilibrium, one finds an exponential distribution, typical of a 1d equilibrium fluid. However, the distribution of separations in the inactive phase is bimodal. For small $d$, the distribution is approximately exponential but with a smaller characteristic length scale than the equilibrium fluid: this corresponds to particles within the dense region of the system. For larger $d$, there is a broad distribution of separations that comes from pairs of particles located on opposite sides of the large void (each configuration contributes only a single sample to the large-$d$ peak, so the width of this peak appears only after averaging many configurations). These separations are comparable to the system size: together with the inhomogeneous density profiles in Fig. 5, they provide quantitative characterization of the phase separation shown in Fig. 4(b).

C. Stability of the phase-separated state

The phenomenon of phase separation is not expected in one-dimensional systems at equilibrium (assuming that forces are short ranged). We therefore explore the physics behind this effect in more detail. One can estimate the probability that the trajectory shown in Fig. 4(b) would occur at equilibrium, as follows. The particles within the large cluster are constrained by their neighbors and their contributions to the activity $K$ are necessarily small. The particles at the boundaries of the cluster cannot move into the cluster, but they may move away from it. The probability that one of these particles nevertheless remains close to the edge of the cluster for the whole time $t_{obs}$ scales as $e^{-\gamma t_{obs}}$, where $\gamma$ is a parameter with units of inverse time, proportional to the rate for diffusion of the particle away from the cluster.

The key point is that maintaining the integrity of the cluster requires only that the two particles at its boundaries do not move away. Hence, for large $t_{obs}$, the probability $P_{ps}$ of a phase-separated state at equilibrium should satisfy $\ln(P_{ps}/P_{eq}) \gtrsim -2\gamma t_{obs}$, where $P_{eq}$ is the probability of a typical equilibrium trajectory. Within the $s$ ensemble, the ratio $(P_{ps}/P_{eq})$ is multiplied by a term $e^{\Delta K}$, where $\Delta K$ is the difference in activity between equilibrium and phase-separated states. Since the activity at equilibrium is extensive (that is, $\Delta K \sim \delta k N l_{obs}$), one therefore expects phase-separated states to dominate for $s \gtrsim 2\gamma/(N \delta k)$.

This argument essentially reproduces the prediction of [33] for phase transitions in kinetically constrained models; see also [13,30]. Assuming that $\gamma$ is independent of $N$, we predict $s^* \lesssim N^{-1}$, consistent with Fig. 3. However, as discussed above, the more refined analysis available from fluctuating hydrodynamics predicts $s^* \sim N^{-2}$ [14]. That is, the bias required to stabilize phase-separated states is even less than that predicted by the simple argument given here. The reason is that the interface between high- and low-density regions of the system may not consist of a single particle, but can be smoothed out: this acts to reduce $\gamma$ to a quantity of order $1/N$, further stabilizing the phase-separated state. However, it seems that the regime in which these smoothed out interfaces can be observed is not accessible within our simulations (where both $N$ and $t_{obs}$ are limited by the computational effort required).

The generalization of these arguments to higher dimensions is not immediate but can we make some tentative predictions. Let $d$ be the spatial dimension and $L$ the linear system size. For phase-separated states in large systems we expect the heuristic argument above to hold, with $\gamma \sim L^{d-1}$ since all particles on the boundary of a large cluster are free to diffuse away. We have $\Delta K \sim L^d$ in all dimensions so we expect a transition to a phase-separated state, with $s^* \lesssim L^{-1}$ in all dimensions. A scaling analysis within fluctuating hydrodynamics yields $s^* \sim L^{-2}$, indicating that the heuristic argument again overestimates $s^*$. In kinetically constrained models [13,30,34], bounds on $s^*$ are available, based on the existence of phase-separated states where almost all particles are unable to move, due to the kinetic constraints. In this case the scaling of $\gamma$ with system size depends on the specific model considered, and can lead to
very small values of $s^*$, as observed (for example) for the two spin facilitated triangular lattice gas model [13] for which we expect $s^* \sim L^{-d}$.

D. Hyperuniformity for $s < 0$

We now turn to the case $s < 0$, in which the system is biased to higher than average activity. In this case, the system enters a hyperuniform state [15], in which density fluctuations on large scales are strongly suppressed. In Fig. 6, we illustrate the density correlations in this state, both in real space and in reciprocal space. Panel (a) shows the pair correlation function $g(x) = \langle \rho(x')\rho(x' + x) \rangle / \langle \rho \rangle^2$ (the system is translationally invariant so there is no dependence on $x'$). Particles appear to repel each other, leading to a depletion zone around each particle, which facilitates motion on small length scales. However, the deviation of $g(x)$ from unity is small in absolute terms: this is a rather weak effect. Moving to Fourier space [Fig. 6(b)] reveals the strong correlations associated with hyperuniformity (similar results were shown in [15]).

In contrast to the equilibrium system for which $S(q) = \text{const}$ for all $q$ [35], one sees behavior consistent with $S(q) \to 0$ as $q \to 0$, which is the signature of hyperuniform states [16]. This transition is predicted to exist in all spatial dimensions; its scaling properties are discussed in [15]. This phenomenon has been found in jammed sphere packings [36,37] and in a range of other physical systems [15,38–40].

IV. RESULTS: CONSTANT PRESSURE

So far, we have discussed the effects of biasing the dynamical activity in the constant-volume hard-particle system, as originally discussed in [15]. We now consider the effects of a similar bias on a constant-pressure system. For consistency with the previous section, we fix the pressure at $P = 7.33(k_B T / l_0)$ so that the mean volume fraction at equilibrium is $\langle \phi \rangle = 0.88$.

A. Inactive state, $s > 0$

The effect of a bias to low activity, $s > 0$, is shown in Figs. 7(a) and 7(b). Comparing with Fig. 2, a similar transition is apparent, but instead of a crossover at some $s^* > 0$ that depends on $N, t_{\text{obs}}$, one instead observes a crossover very close to the equilibrium point $s = 0$. Note that the values of $t_{\text{obs}}$ used here are significantly larger than those used in the constant-volume system; they are comparable with the volume

FIG. 6. (Color online) (a) The pair correlation function, $g(x)$, for point particles in the constant-density regime with $N = 120$, $t_{\text{obs}} = 20\tau_L$ in the active phase. The depression in $g(x)$ for small $x$ reflects an increase in local free volume around the particles as activity increases. (b) Small $q$ structure factor in constant-density systems. Biasing to $s < 0$ causes the suppression at small wave vectors, $S(q) \sim q^2$, consistent with the onset of hyperuniformity. At fixed $s$ all system sizes collapse onto a single curve.

FIG. 7. (Color online) Dynamic behavior of the constant-pressure system. All results come from trajectories of duration $t_{\text{obs}} \approx 2\tau_L$ at $P = 7.33(k_B T / l_0)$. (a) The intensive activity and dynamic susceptibility of the system as a function of $s$. The transition (crossover) takes place at $s^* = 0$ for all system sizes, and the width of the crossover is proportional to $(\tau_L)^{-1}$. (b) The data for all system sizes collapse when scaled by $\tau_L$; inset: the peak in the dynamic susceptibility scales with $\tau_L$. (c) The (total) density of the system changes as the system undergoes the transition shown in (a).
Given this strong correlation, we can link the phase transition that takes place at $s = 0$ in this system to the hydrodynamic time scale $\tau_{LL}$, which diverges in the limit of large systems (recall Sec. II A 2). We define the (normalized) autocorrelation function of the system size

$$C_{LL}(t) = \frac{\langle \delta L(t') \delta L(t' + t) \rangle_{eq}}{\langle \delta L(t')^2 \rangle_{eq}},$$

which is evaluated at equilibrium (so there is no dependence on $t'$), with $\delta L = L - \bar{L}$. In the low-pressure limit, this correlation function decays as $e^{-t/\tau_{LL}}$. For the finite pressures considered here, the correlation time is also close to $\tau_{LL}$. Similarly the correlation function of the activity is

$$C_{kk}(t) = \frac{\langle \delta K(t') \delta K(t' + t) \rangle_{eq}}{\langle \delta K(t')^2 \rangle_{eq}},$$

where $K(t) = \sum_i [r_i(t + \Delta t) - r_i(t) - \Delta \bar{L}(t)]^2$ is the quantity that appears in the definition of the activity $K$; recall (9). To show the long-time behavior of $C_{kk}(t)$ more clearly we smooth the function by convolving it with a Gaussian window, with variance $\sigma^2 = \tau_{LL}^2/4$: we plot $\Gamma \sum_i C_{kk}(t') e^{-2(t-t')^2/\sigma^2}$, where the proportionality constant $\Gamma$ normalizes the correlation function to unity at $t = 0$.

Figure 10 shows that the correlation functions $C_{kk}(t)$ and $C_{LL}(t)$ behave very similarly, consistent with the idea that the activity fluctuations are strongly correlated with those of the global density (and hence to the system size). Since the relaxation time of the barostat $\tau_{LL} \sim \bar{T}^2$ (recall Sec. II A 2).

The density of the system also shows a sharp crossover near $s = 0$; see Fig. 7(c).

Figure 8 shows representative trajectories from biased ensembles in the constant-pressure system. Comparing with Fig. 4, no phase separation occurs. We also note that the box size varies with $s$, consistent with Fig. 7(c).

Figure 9 shows the correlation between activity and average density for all dynamic regimes. Within the theory of fluctuating hydrodynamics [17,18], the density field is assumed to give a full description of the large-scale behavior of this system. In constant-volume systems, density fluctuations on finite wave vectors control the fluctuations in activity [15,27]. In the constant pressure system, Fig. 9 shows that the total density (i.e., the density at zero wave vector) correlates strongly with the activity: high density is associated with low activity, and vice versa.

**FIG. 8.** (Color online) Representative trajectories of constant pressure systems at $P = 7.33(k_B T / \bar{L})$ with $N = 40, t_{obs} = 220\tau_0$ at different biases. Red lines indicate the boundaries of the system and dashed lines show the mean box size at equilibrium. (a) Active state, $s = -0.25$; the system expands, reducing its density and leading to increased activity. (b) $s = 0$, the equilibrium system has a fluctuating volume but maintains increased activity. (c) Inactive state, $s = 0.375$: the system size is reduced relative to equilibrium, suppressing the activity.

**FIG. 9.** (Color online) Scatter plot of activity against the average density of trajectories of duration $1300\tau_0$ with $N = 100$ particles from biased ensembles. There is a strong correlation between the activity and the density of the trajectories. The data come from ensembles with a range of bias strengths, as shown, and cover a wide range of density and activity. The gaps between data sets are due to the limited set of bias values for which we show data: trajectories do occur with all values of $\phi$ and $k$.

**FIG. 10.** (Color online) The activity and volume correlation functions for constant pressure systems at equilibrium, for different system sizes. The correlation time $\tau_{LL}$ is strongly correlated with $\tau_{LL}$. To display the long-time behavior most clearly, $C_{kk}(t)$ has been smoothed with a Gaussian window (see main text).
volume relaxation time \( \tau_{LL} \) diverges as \( \frac{1}{N} \), we therefore expect a similar divergence in the relaxation time of the activity.

This divergent time scale is important because the susceptibility \( \chi = -dk/ds \) is related to the autocorrelation function of the activity [30] as

\[
\chi(s = 0, t_{\text{obs}} \to \infty) = \frac{2}{L} \int_0^\infty dt \, \langle \delta K(0) \delta K(t) \rangle_{\text{eq}}
\]  

so that \( \chi \sim \tau_{kk} \sim \frac{1}{L} \) diverges at \( s = 0 \), which we interpret as a dynamical phase transition. (The equal time value of the correlator in this equation scales as \( L \) since \( \delta K \) is extensive in the system size: this \( L \) dependence cancels with the prefactor so that the right hand side scales with \( \tau_{kk} \), with a prefactor of order unity.) Since \( \chi = -dk(s)/ds \), the divergence of \( \chi \) corresponds to a singularity in \( k(s) \) and hence a dynamical phase transition. This amounts to a perturbative argument for the existence of the phase transition: a related perturbative argument based on fluctuations at finite wave vector was used in [15] to explain the existence of phase transitions in systems at finite volume.

Recalling the discussion of Sec. III C, the analogous argument for the constant-pressure system is that the noise force \( \eta_L \) in (6) acquires a finite average in the inactive state, resulting in a reduced system size. The previous argument based on \( \tau_{LL} \) indicates that biasing the noise force in this way requires very little cost in probability: this low cost appears partly because only a single noise term needs to be biased, but also because the absolute size of the bias becomes small in large systems, due to the scaling of the diffusion constant \( D_L \) with system size [Eq. (8)].

Finally we note that \( s^* \) shifts from a value of order \( 1/N \) in the constant-density system to a value close to zero at constant pressure. We interpret the small positive \( s^* \) (for the constant-volume system) in terms of the probability cost required to form the interface in a phase-separated system at constant density. At constant pressure, no interface is required so the system can have a diverging linear response, as shown in (17).

**B. Active state, \( s < 0 \)**

In Fig. 11(a), we show the structure factor of the constant-pressure system for \( s < 0 \). For a given bias \( s \), the fluctuations in the total system size are small in relative terms, so we evaluate the structure factor at wave vectors \( q = 2n\pi/L' \) as usual, and calculate \( S(q) \) by an ensemble average at fixed \( n \) (recall \( L' = L - N\ell_0 \) is the system size in the representation where particles are treated as pointlike). This provides an estimate of \( S(q) \) with \( q = 2n\pi/L' \). The results of Fig. 11(a) are consistent with hyperuniformity of the active \( (s < 0) \) phase, although the effect is weaker than that shown in Fig. 6, for the constant-density system. We also show the distribution of particle separations in Fig. 11(b), for comparison with Fig. 5(b). The distribution fits well to an exponential form, independent of \( s \). Given the correlations that are apparent from Fig. 11(a), this result is somewhat surprising: it might be that the correlations are sufficiently weak on short length scales that they are not discernible from \( P(d) \).

In this constant-pressure system, it seems that achieving large deviations by changes in structure (for example phase separation or hyperuniformity) is unfavorable compared to changing the system density. Thus, particle separations remain exponentially distributed independent of \( s \), but the system density depends strongly on \( s \). Even at the longest length scales and largest \( |s| \) the structure is only weakly affected; compare Figs. 6(a) and 11(a).

It is possible that a more accurate mimic of the active constant-volume system would be obtained by a constant-pressure system whose pressure was adjusted to match the (fixed) density of the constant-volume one. This would be consistent with the idea that the (virial) pressure in the constant-volume system depends on the bias \( s \). We defer an investigation of this possibility to a future study.

**V. DISCUSSION: PRESSURE BALANCE AND MECHANICAL EQUILIBRIUM**

In Secs. III and IV, we have presented numerical results for different biases and in different ensembles. To rationalize these results, it is useful to consider the fundamental principles of force balance and mechanical equilibrium in these biased (nonequilibrium) ensembles. Of particular interest is the
The forces acting in this situation are analyzed in Appendix B, under the assumptions that there are no macroscopic particle currents in the steady state of the system, and that the forces at the boundaries come only from the conservative forces between particles. (We neglect any forces from momentum fluxes across the boundaries.) The analysis follows the standard methodology for equilibrium systems [41] (see also [42]). Here we state the main results.

In the steady state, balance of the total forces on the subsystem yields \( \nabla P_{\text{mech}} \) of the subsystem (which may have nonzero averages if \( s \neq 0 \)). The forces acting in this situation are analyzed in Appendix B, under the assumptions that there are no macroscopic particle currents in the steady state of the system, and that the forces at the boundaries come only from the conservative forces between particles. (We neglect any forces from momentum fluxes across the boundaries.) The analysis follows the standard methodology for equilibrium systems [41] (see also [42]). Here we state the main results.

In the steady state, balance of the total forces on the subsystem yields \( \nabla P_{\text{mech}} \) of the subsystem (which may have nonzero averages if \( s \neq 0 \)).
exerted on a subsystem, and we assume (i) that no macroscopic currents flow in the biased ensemble, and (ii) that the pressure is isotropic everywhere, so that any forces on a subsystem are orthogonal to its boundaries. The possible relationship between these calculations and active-matter systems [20–23] is an interesting direction for future study.

VI. CONCLUSION

We have demonstrated dynamic phase transitions in a one-dimensional model of diffusing particles, including transitions from simple equilibrium fluid states into both high-activity and low-activity states. We considered both constant-density and constant-pressure systems: their transitions share some common features but there are also important differences. Based on the theory of fluctuating hydrodynamics, we argued [15] that the transitions occur for all densities \( \rho \), and the arguments given here indicate that this should also hold for all applied pressures.

Considering first the transition to inactive states, the constant-volume system undergoes phase separation, while the constant-pressure system increases the local density. (As in equilibrium systems, the constant-pressure system avoids interfaces between coexisting phases.) For large systems, the inactive phase occurs for all \( s > 0 \), in both ensembles. However, there is no signature of phase coexistence in the constant-pressure system: we find only a dense phase, consistent with the different (mechanical) pressures of the coexisting phases in the constant-volume system.

For transitions to high-activity states, the constant-volume system spontaneously suppresses long-range density fluctuations and develops a hyperuniform structure [15]. At constant pressure, the main feature of the high-activity state is that the total density decreases sharply, although there is also some suppression of large-scale density fluctuations.

Overall, these results emphasize that equilibrium ideas of ensemble equivalence do not apply directly when considering large deviation phenomena such as those considered here. While the mechanical pressure and the virial can still be related, the possibility of phase coexistence at unequal pressures shows how familiar equilibrium concepts such as phase separation need to be re-evaluated and generalized in these nonequilibrium settings.

The supporting data for this article are openly available from the University of Bath data archive [44].

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APPENDIX A: MONTE CARLO DYNAMICAL SCHEME

1. Constant volume system

The constant-volume system evolves by single particle MC moves [19]. Particle displacements \( \Delta x \) are chosen uniformly from the range \(-S \leq \Delta x \leq S\), with \( S = 0.1 l_0 \). If the displacement results in a particle overlap, it is rejected; otherwise, it is accepted.

For small time steps and on short time scales (in between collisions), particles undergo diffuse motional: a particles’ mean-squared displacement after \( N_{MC} \) steps is \( N_{MC} S^2/3 \). From the Langevin equation (1), we note that the displacement of a particle after a similarly short time \( t \) is \( 2D \pi t \). For consistency between Langevin and MC descriptions, we equate these displacements, which shows that the physical time associated with one MC move (per particle) is \( t_0 = S^2/(6D \pi) \).

When the packing fraction \( \phi \) is large, the rate of acceptance of MC moves can get small, and it becomes convenient to use a rejection-free MC algorithm that operates in continuous time \([19,45]\). To achieve this, all possible particle displacements are calculated: let \( g_i \) denote the fraction of possible moves for particle \( i \) that are compatible with the hard-particle interactions (i.e., the fraction that would be accepted). Particle \( i \) is selected with probability \( g_i/(\sum j g_j) \) and one of its possible moves is implemented. The simulation time is then incremented by

\[
\tau_i = \frac{\tau_B}{N_{MC}} \frac{N}{\sum j g_j} \ln(1/\mu),
\]

where \( \mu \) is randomly distributed \( 0 < \mu \leq 1 \) [so \( \ln(1/\mu) \) is exponentially distributed with a mean of unity]. The process is repeated and moves are made until the total simulation time reaches the desired duration.

2. Constant pressure system

In constant-pressure simulations, the particle coordinates evolve as above. In addition, we also perform MC moves in which we propose changes to the volume of the system. A change in volume is proposed as \( L_{new} = L_{old} + \Delta L \) where \(-S_L \leq \Delta L \leq S_L\). The scaled particle positions \( u_i \) are the same in the old and new configurations. If the proposed move causes any particles to overlap the move is rejected immediately. Otherwise, the move is accepted with probability \( \min(1,e^{-\beta \Delta L + N \ln L_{new}/L_{old}}) \) [19].

At every MC step, a volume change is proposed with fixed probability \( 1/N \) and a particle displacement is proposed with probability \( (N - 1)/N \). The scheme samples the correct equilibrium distribution independent of this probability and independent of the move size \( S_L \). However, to ensure consistency with Eqs. (6) and (8) we specify that the maximum volume change satisfies \( S_L^2 = S^2(2\pi)^2 k_B T \kappa /L \). (Note this depends on the applied pressure, via the mean box size \( L \).

APPENDIX B: FORCE BALANCE IN BIASED ENSEMBLES

In this appendix, we consider the forces acting on the subsystem shown in Fig. 12, and we derive a formula for the pressure of this subsystem.

1. Force balance

We first consider force balance. Starting from the equation of motion (1), we sum over all particles inside subsystem \( B \). Taking an average within a (possibly inhomogeneous) steady state and assuming no net currents we have \( \sum_i L \dot{x}_i = 0 \). It is convenient to define \( \xi_i = \eta_i k_B T \sqrt{2/m_D} \), which is the force associated with the noise term \( \eta_i \) in the equation of motion (1).
Hence from (1),

\[ \sum_{i \in B} \sum_{j \text{ outside } B} F_{ij} + \sum_{i \in B} \xi_i = 0, \quad (B1) \]

where \( F_{ij} = -F_{ji} \) is the conservative (nonstochastic) force exerted on particle \( i \) by particle \( j \); in our system \( F_{ij} = \psi(x_i - x_j) \). Here, \( \psi(x) \) is the interparticle potential [recall (3)] and the prime denotes a derivative. We use an overbar instead of angle brackets to indicate averages within steady states, to avoid possible confusion if ergodicity is broken. For example, if phase separation occurs as in Fig. 4, the steady-state average is taken with a fixed (arbitrary) position of the void, while an ensemble average (indicated here by angle brackets) should include all possible void positions.

The first term (double sum) in (B1) is localized at the boundary of the subsystem. Recalling Fig. 12, we decompose the sum into contributions from the different boundaries. Assuming that the total force on each boundary is normal to that boundary, we take the \( x \) component of the forces in (B1). The double sum reduces to \(-F^{xx}(X_R) + F^{xx}(XL)\), since \( F^{xx} \) is defined as the sum over the boundary of the forces acting on particles inside the system. Hence (B1) yields

\[ -F^{xx}(X_R) + F^{xx}(XL) + \overline{\xi} = 0, \quad (B2) \]

where \( \overline{\xi} = \sum_{i \in B} \xi_i \) is the \( x \) component of the total noise force. We emphasize that the \( F^{xx} \) are sums of conservative interparticle forces only.

Recalling \( p^{\text{mech}}(X) = F^{xx}(X)/A_1 \), where \( A_1 \) is the area of the boundary, we find

\[ \overline{F}^x = \frac{p^{\text{mech}}(X_R) - p^{\text{mech}}(XL)}{X_R - XL}, \quad (B3) \]

where we used \( V_B = (X_R - XL)A_1 \). This equation states that any average noise forces on the subsystem must be balanced by conservative forces that act at the subsystem boundaries. We further assume that the pressure drop across the subsystem is small enough to write \( p^{\text{mech}}(X_R) - p^{\text{mech}}(XL) \approx (X_R - XL)(\partial p^{\text{mech}}/\partial x) \). Considering the other boundaries in a similar way, one arrives at Eq. (18) of the main text.

2. Virial expression for the pressure

We now follow a standard analysis that relates the pressure in a system to the virial [41]. We require a mild generalization in order to include the possibility that noise forces in the Langevin equation have finite averages—a similar situation is considered in [42], for active matter systems. It is useful to refer to Fig. 12. We define the virial for the entire system by

\[ V = \frac{1}{2d} \sum_{ij} (x_i - x_j) F_{ij}, \quad (B4) \]

which includes only the conservative forces.

It is useful to decompose \( V \) into four contributions:

\[ V = V_B + V_{AB} + V_{BA} + V_A, \quad (B5) \]

Here \( V_B = \frac{1}{d} \sum_{i \in B} \sum_{j \text{ outside } B} (x_i - x_j) F_{ij} \) [recall Eq. (19)] is the contribution from particles entirely inside \( B \), while

\[ V_{AB} = \frac{1}{d} \sum_{i \in B} \sum_{j \text{ outside } B} x_i F_{ij} \]

involves particles near the boundary of \( B \). The term \( V_{BA} = \frac{1}{d} \sum_{i \text{ outside } B} \sum_{j \in B} x_j F_{ij} \) is also localized at the interface, while \( V_A \) involves contributions only from particles outside region \( B \). (We include factors of \( d \) in these definitions for later convenience.)

The next step is to relate the mechanical pressure to \( V_{AB} \), following [41, 42]. We decompose (B6) into a sum over the different boundaries of system \( B \), and decompose each \( F_{ij} \) into components parallel and normal to the boundary. For concreteness, consider the rightmost boundary in Fig. 12. All particles at the boundary have \( x_i \approx X_R \), so the contribution of this boundary to \( V_{AB} \) is given by (1/d) \( \sum_{i \in B} \sum_{j \text{ outside } B} x_i F_{ij} \approx (X_R/d) \sum_{j \text{ outside } B} \sum_{i \in B} F_{ij} \), where the sums run over particles \( i \) that are inside \( B \) and close to the relevant boundary, and particles \( j \) that are outside \( B \). The second equality comes from the definition of \( F^{xx}(X_R) \) as the sum of conservative forces at the boundary. Similarly the leftmost boundary gives a contribution \( X_L F^{xx}(XL)/d \) to \( \overline{V_{AB}} \). The sum of these contributions is

\[ V_{AB} = -\frac{1}{2d} (X_R - XL)[F^{xx}(X_R) + F^{xx}(XL)] - \frac{1}{2d}(X_R + XL)(F^{xx}(X_R) - F^{xx}(XL)). \quad (B7) \]

where the superscript \( x \) indicates that we considered only interfaces perpendicular to the \( x \) axis. Assuming as before that these quantities vary weakly on the scale of the subsystem, a Taylor expansion of \( F^{xx} \) yields

\[ V_{AB} \approx -\frac{1}{d} (X_R - XL)[F^{xx}(X_R) + X_R(\partial F^{xx}/\partial x)] \]

\[ -\frac{1}{d}(X_R + XL)(F^{xx}(X_R) - F^{xx}(XL)). \quad (B8) \]

with \( X_B = (X_R + XL)/2 \) the geometrical center of the subsystem. [We have neglected corrections at \( O(X_R - X_L)^2 . \]

In one spatial dimension, \( V_{AB} \) is given by (B8). In higher dimensions, the virial has other similar contributions that come from the other pairs of parallel interfaces. Incorporating these terms and recalling from Fig. 12 that \( p^{\text{mech}} = F^{xx}/A_1 \), the generalization of (B8) is

\[ \frac{V_{AB}}{V_B} = -X_B \cdot \nabla p^{\text{mech}}(X_B). \quad (B9) \]

where we used \( A_1(X_R - XL) = V_B \). Alternatively using (18) we may write

\[ V_{AB} = -p^{\text{mech}}(X_B)V_B - X_B \cdot \sum_{i \in B} \xi_i. \quad (B10) \]

(Recall that \( \xi_i = \eta_i k_B T \sqrt{2/D_p} \) is the force corresponding to the stochastic noise \( \eta_i ).

The final step is to obtain a formula for the local pressure that depends only on particles inside \( B \). To this end, we use the definitions of the virials to write \( V_{AB} + V_{BA} = (1/d) \sum_{i \in B} \sum_{j \text{ outside } B} x_i F_{ij} = (1/d) \sum_{j \text{ outside } B} x_j F_i \), where \( F_i = \sum_j F_{ij} \) is the total conservative force on particle \( i \). From the equation
of motion (1) we have therefore

\[ \mathcal{V}_B + \mathcal{V}_{AB} = \frac{1}{d} \sum_{i \in B} x_i \left( \frac{k_B T}{D_p} \partial_x x_i - \xi_i \right) \]

\[ = \frac{1}{d} \sum_{i \in B} \left[ \frac{k_B T}{D_p} \partial_t (x_i^2/2) - d k_B T - x_i \xi_i \right], \]

(B11)

where we used Ito's formula \( \partial_t f(x_i) = f'(x_i) \partial_x x_i + f''(x_i) D_p \).

The analysis at this point is slightly subtle (see also [42]). The simplest approach is to assume that the average of the time derivative \( \partial_t (x_i^2) = 0 \) in the steady state, so we obtain \( \overline{\mathcal{V}_B} + \overline{\mathcal{V}_{AB}} = -N_B k_B T - \frac{1}{d} \sum_{i \in B} x_i \xi_i \), where \( N_B \) is the number of particles in the subsystem. Using (B10) to substitute for \( \mathcal{V}_{AB} \), one finally arrives at

\[ p^\text{mech}(x_B) \mathcal{V}_B = N_B k_B T + \overline{\mathcal{V}_B} + \frac{1}{d} \sum_{i \in B} \xi_i (x_i - x_B) \]  

(B12)

which is equivalent to (21) of the main text (note \( \delta x_i = x_i - x_B \)). The terms proportional to \( N_B \) and \( \mathcal{V}_B \) are familiar from equilibrium [41] but the final "swim pressure" term is new [23]. Thus Eq. (B11) allows evaluation of the mechanical pressure from knowledge of the positions and noise forces of particles inside subsystem B; see also Eq. (20) of the main text.

Strictly, the assumption that \( \partial_t (x_i^2) = 0 \) can be justified only if particles in \( B \) are confined within its boundaries, so that they equilibrate to a constant density profile. For a more precise analysis [42], it is necessary to consider particles that exchange between regions \( A \) and \( B \) of the system, and the associated momentum fluxes. This case will be analyzed in a future publication.