Hyperuniformity and Phase Separation in Biased Ensembles of Trajectories for Diffusive Systems

Robert L. Jack,[1] Ian R. Thompson,[1] and Peter Sollich[2]
[1]Department of Physics, University of Bath, Bath BA2 7AY, United Kingdom
[2]Department of Mathematics, King's College London, Strand, London WC2R 2LS, United Kingdom
(Received 23 September 2014; published 9 February 2015)

We analyze biased ensembles of trajectories for diffusive systems. In trajectories biased either by the total activity or the total current, we use fluctuating hydrodynamics to show that these systems exhibit phase transitions into "hyperuniform" states, where large-wavelength density fluctuations are strongly suppressed. We illustrate this behavior numerically for a system of hard particles in one dimension and we discuss how it appears in simple exclusion processes. We argue that these diffusive systems generically respond very strongly to any nonzero bias, so that homogeneous states with "normal" fluctuations (finite compressibility) exist only when the bias is very weak.

DOI: 10.1103/PhysRevLett.114.060601 PACS numbers: 05.40.-a

Introduction.—Nonequilibrium systems exhibit diverse collective behavior and complex emergent phenomena, many of which have no counterparts at equilibrium. Even in simple interacting particle systems, one may encounter long-ranged correlations [1], dissipative "avalanche" events with no typical size [2], and dynamical phase transitions [3,4]. Theories that capture the universal aspects of these fluctuations are much sought after, as a route to general descriptions of nonequilibrium phenomena. Here, we analyze nonequilibrium ensembles of trajectories [3–5], defined through constraints on macroscopic observables such as the total current or activity within a given time period. Phase transitions within these ensembles occur when such a constraint leads to a qualitative change in macroscopic behavior [3,4,6,7]. In diffusive systems [8–12], we demonstrate transitions into "hyperuniform" states [13], as well as transitions into the macroscopically inhomogeneous ("phase-separated") states found previously [3,6]. Hyperuniform states are characterized by anomalously small density fluctuations on large length scales [13–20]; they have been identified in jammed particle packings [16,17] and in biological systems [19]. These systems are highly optimized in response to a global constraint (mechanical stability in jamming, optimal fitness in biology). The constrained dynamical ensembles considered here are also optimized: they are the maximally probable states consistent with the constraint. Our results (i) provide further evidence that hyperuniformity is generic, by demonstrating that it occurs in a new set of optimized nonequilibrium ensembles, and (ii) resolve the physical interpretation of some phase transitions that have been previously discovered in diffusive systems [6,21].

Models.—We study biased ensembles of trajectories both computationally and analytically. For computational studies, we consider a one-dimensional model of N diffusing hard particles of size $l_0 = 1$, in a periodic box of size L. This Brownian hard-particle model evolves by a dynamically realistic Monte Carlo scheme: for small time steps, we recover motion consistent with overdamped Langevin dynamics. The diffusion constant of an isolated free particle is D_0. The hard particles may not overtake one another, which means that the system may be mapped to a system of point particles. Full system details, including this mapping, are given in the Supplemental Material (SM) [22].

We also consider lattice-based exclusion models where N particles are distributed over L lattice sites, again with periodic boundaries. At most, one particle may occupy any lattice site. In the (partially) asymmetric simple exclusion process (ASEP), particles hop left with rate ℓ and right with rate r, provided their destination site is empty. The symmetric simple exclusion process (SSEP) is the case $\ell = r = 1$. All of the models considered here have trivial steady-state correlations, in which particles are independently distributed, subject to the exclusion constraints. The mean density is $\bar{\rho} = N/L$.

Biased ensembles of trajectories.—Let $K = K[x(t)]$ be a measure of dynamical activity in a trajectory $x(t)$. For exclusion processes, K is the total number of particle hops in a trajectory. For the Brownian hard-particle model, we define a coarse-graining time $\tau_0 = \ell_0^2/(2D_0)$, the time for an isolated a particle to diffuse a distance comparable with its size. We focus on trajectories of length $t_{\rm obs} = M\tau_0$, defining $K = \sum_{j=1}^{M}\sum_{i=1}^{N}|\hat{x}_i(t_j) - \hat{x}_i(t_{j-1})|^2$ with $t_j = j\tau_0$ [7]. The position $\hat{x}$ is defined by subtracting a kind of center-of-mass motion [22], which helps to minimize finite-size effects. The unit of time is $\tau_0 = 1$.

To investigate trajectories with nontypical values of K, we define a biased ensemble of trajectories [4,5,27], via a formula for the average of an observable O:

$$\langle O\rangle_s = e^{-\psi_K(s)L^d t_{\rm obs}}\langle O e^{-sK}\rangle_0. \quad (1)$$

Here, $\langle\cdot\rangle_0$ represents an average in the (unbiased) steady state of the model, $\langle O\rangle_s$ is an average within the biased ensemble, and $\psi_K(s) = \log\langle e^{-sK}\rangle_0/(L^d t_{\mathrm{obs}})$ is a "dynamical free energy." For sufficiently large t_{obs}, averages in the biased ensemble are equal to averages over trajectories in which the activity K is constrained [28].

Numerical results.—Figure 1 shows results for the Brownian hard-particle model, calculated using transition path sampling [7,29]. Figure 1(a) shows the mean activity $k(s) = (L^d t_{\mathrm{obs}})^{-1}\langle K\rangle_s$. For $s > 0$, there is a first-order transition into an (inhomogeneous) phase-separated state [3,6,30], as shown in the inset. For $s < 0$, the activity appears to depend smoothly on s, but the system develops strong long-ranged correlations. These are measured by the structure factor $S(q) = (1/L^d)\langle\delta\rho_q(t)\delta\rho_{-q}(t)\rangle$ where $\delta\rho_q = \int dr\delta\rho(r)e^{-iq\cdot r}$ and $\delta\rho(r) = \rho(r) - \bar{\rho}$. To see the relevant behavior most clearly, we transform coordinates so that the particles are treated as pointlike [22]: the equilibrium ($s = 0$) ensemble then has $S(q) = N/(L - Nl_0)$, independent of q. Figure 1(b) shows that for $s < 0$ and small q, the structure factor deviates strongly from this equilibrium value. The signature of a hyperuniform state is that $S(q) \sim q$ at small q [13]: density fluctuations on large length scales are strongly suppressed. This means that particle positions necessarily have long-ranged correlations [otherwise, self-averaging of the density implies $\lim_{q\to 0} S(q) > 0$]. Analysis of the small-q behavior in numerical simulations is limited by the system size, but the results for $s < 0$ are consistent with hyperuniformity. All results were obtained at density $\bar{\rho} = 0.88$: the following analysis indicates that the qualitative behavior of this system as $L \to \infty$ is independent of $\bar{\rho}$, although numerical factors and finite-size effects will depend on this parameter.

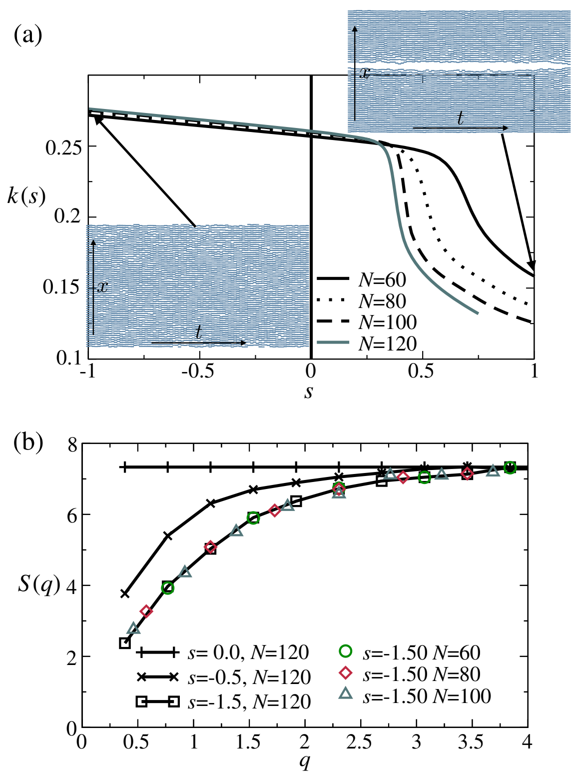

FIG. 1 (color online). Numerical results for the Brownian hard-particle model. (a) Mean activity $k(s)$ in biased ensembles at $\bar{\rho} = 0.88$, with $t_{\mathrm{obs}} = 20\tau_0$. The insets illustrate representative trajectories at $s = \pm 1$, in which particles' world lines (blue regions) are visualized in space and time. For $s > 0$, the system phase separates and a macroscopic region of empty space appears, accompanied by a jump in $k(s)$; for $s < 0$, the system is homogeneous. (b) Structure factor $S(q)$ in biased ensembles. For $s < 0$, small-q density fluctuations are strongly suppressed.

Fluctuating hydrodynamics and hyperuniformity.—The models considered here fall into a general class of diffusive systems, which may be described by "fluctuating hydrodynamics" [1,12,31]. Within this theory, the time evolution of the density $\rho(r, t)$ on large length and time scales can be described by a Langevin equation

$$\partial_t\rho(r,t) = \nabla\cdot D[\nabla\rho(r,t) - a] + \nabla\cdot[\sqrt{\sigma}\eta(r,t)], \quad (2)$$

where η is a white noise, $D = D[\rho(r,t)]$ and $\sigma = \sigma[\rho(r,t)]$ are local measures of diffusivity and mobility, and a is an asymmetric driving force. Details of the relationships between fluctuating hydrodynamics and the models considered here are given in the SM [22]. The fluctuating hydrodynamic theory is valid in all dimensions, not just $d = 1$.

We now consider a bias to larger-than-average activity $s < 0$, applied to a system described by (2), with $a = 0$. Averages within the biased ensemble are given by path integrals: $\langle O\rangle_s = e^{-\psi_K(s)L^d t_{\mathrm{obs}}}\int\mathcal{D}\rho\mathcal{D}\hat{\rho}O[\rho]e^{-\int drdt\mathcal{L}}$, where $\hat{\rho}$ is a response field, and

$$\mathcal{L} = i\hat{\rho}[\partial_t\rho - \nabla\cdot(D\nabla\rho)] + \frac{1}{2}\sigma(\nabla\hat{\rho})^2 + s\kappa, \quad (3)$$

in which $\kappa = \kappa(\rho)$ is the (density-dependent) local activity of the system. We assume $\kappa''(\rho) \le 0$, which certainly holds for exclusion processes and may be expected for generic particle systems; analyzing the case with $\kappa''(\rho) > 0$ is also straightforward [32–34]. The behavior of $\kappa(\rho)$ for the Brownian hard-particle model is shown in the SM [22].

Analysis of hydrodynamic behavior requires a suitable rescaling of space and time coordinates. To avoid cumbersome notation, we defer this procedure to the SM [22] and quote our results in terms of the bare (unrescaled) parameters. Note, however, that these results apply only in the hydrodynamic limit. For $s \le 0$, the path integral is dominated by trajectories where $\rho(r,t) \approx \bar{\rho}$ and $\hat{\rho} \approx 0$, so we write $\rho(r,t) = \bar{\rho} + \delta\rho(r,t)$ and expand to quadratic order in $\delta\rho$ and $\hat{\rho}$. We also expand $\kappa(\rho) = \kappa_0 + \kappa_0'\delta\rho + \kappa_0''\delta\rho^2/2 + \cdots$, with $\kappa_0 = \kappa(\bar{\rho})$, $\kappa_0' = (d/d\rho)\kappa(\bar{\rho})$, etc., and similarly for $D(\rho)$ and $\sigma(\rho)$. The structure factor may then be evaluated (see Eq. (58) of Ref. [6] and also the SM [22]), yielding

$$S(q) = \frac{\sigma_0 q^2}{2\sqrt{(D_0 q^2)^2 + s q^2 \sigma_0 \kappa_0''}}. \tag{4}$$

This result applies only on the hydrodynamic scale: it should be valid for arbitrary values of the ratio q^2/s, but we expect corrections at $O(s)$ and $O(q^2)$. Recall that $s, \kappa_0'' \leq 0$, by assumption.

The case $\kappa_0'' = 0$ corresponds to completely noninteracting particles, in which case the bias s has no effect on the structure. However, for any $\kappa_0'' < 0$, Eq. (4) demonstrates a singular response to the field s. For $s = 0$ and $q \to 0$, the structure factor approaches a nonzero constant $\sigma_0/(2D_0)$, as expected in an equilibrium state with a finite compressibility. However, for any $s < 0$, the large-scale behavior changes qualitatively: $S(q) = (q/2)\sqrt{\sigma_0/(s\kappa_0'')} + O(q^2/s)$, which is consistent with the numerical results of Fig. 1(b). Note that hyperuniformity is a large length scale phenomenon: the nontrivial behavior in $S(q)$ appears only for small $q \lesssim \sqrt{s\sigma_0\kappa_0''}/D_0$.

We also calculate the mean activity $k(s) = \langle\kappa\rangle_s \approx \kappa_0 + (\kappa_0''/2)\langle\delta\rho(r,t)^2\rangle_s$. Writing $\langle\delta\rho(r,t)^2\rangle = (2\pi)^{-d}\int d^d q S(q)$, we see that the suppression of $S(q)$ at small q acts to increase $k(s)$ (recall $\kappa_0'' < 0$). Taking $s < 0$ and $d = 1$, we obtain [21,22]

$$k(s) - k(s=0) \approx \sqrt{s\kappa_0''\sigma_0}\frac{|\kappa_0''|\sigma_0}{4\pi D_0^2}, \tag{5}$$

which is valid to leading order in $|s|$. Since $k(s) = -\psi_K'(s)$ where ψ_K is the dynamical free energy, we identify this nonanalytic behavior in $k(s)$ with a second-order dynamical phase transition. This singular behavior has been noted before [21], but its link with hyperuniformity has not. In $d > 1$, the suppression of $S(q)$ at small wave vectors leads to a singular contribution $k(s) - k(s=0) \sim (-s)^{d/2}$, with logarithmic corrections if d is even [22]. The predicted singular behavior in $k(s)$ requires a joint limit of large L and large t_{obs}: in Fig. 1, these effects are smoothed out by finite-size (and finite-t_{obs}) effects. In particular, the singular behavior predicted by (5) is not readily apparent: we expect the divergence of $k'(s)$ as $s \to 0^-$ to become visible only for significantly larger t_{obs} [35]. Biasing to lower-than-average activity ($s > 0$, rather than $s < 0$ as so far) results in a macroscopically inhomogeneous (phase-separated) state [3,6,30,32] as illustrated in Fig. 1. Finite-size scaling analysis also predicts that the bias s^* required to cause phase separation scales as L^{-2} [30], consistent with the trend observed in Fig. 1.

Heuristic arguments for hyperuniformity.—The origin of hyperuniformity in these systems is the diverging hydrodynamic time scale associated with large-scale density fluctuations. To see this, consider linear response to the field s. Within a biased ensemble of trajectories, the probability of finding the system in configuration $\mathcal{C}$ is $p_{\mathcal{C}}(s) = p_{\mathcal{C}}(0)[1 - 2s\int drdt\langle\delta\kappa(r,t)\rangle_{\mathcal{C}} + O(s^2)]$ where $\langle\delta\kappa(r,t)\rangle_{\mathcal{C}}$ is a "propensity" [36], obtained by averaging the activity over trajectories that start in $\mathcal{C}$ at $t = 0$, and comparing with typical equilibrium trajectories [22,37,38].

If $\mathcal{C}$ has an unusual density fluctuation at a small wave vector $q \approx 1/R$, expanding $\delta\kappa$ to quadratic order in $\delta\rho$ gives $\int dr\delta\kappa(r,t) \approx (\kappa_0''/2L^d)[|\rho_q(t)|^2 - \langle|\rho_q(t)|^2\rangle_0]$. Diffusive scaling indicates that these correlations relax on a time scale $\tau_R = R^2/D_0$ which diverges for large R: this leads to a corresponding divergence in the linear responses $p_{\mathcal{C}}'(s)$, due to the time integral of $\delta\kappa$. For $s < 0$, it is the hyperuniform states that receive the strongest enhancement [22]; a similar effect appears for phase-separated states if $s > 0$. Similar links between diverging relaxation times and dynamical phase transitions are found in glassy systems [37,39].

Biased ensembles based on the total current.—So far, we have considered ensembles of trajectories biased according to their activity K. In fact, hyperuniform states also appear in ensembles of trajectories where the total current is biased. Here, the total current J is the sum of all (directed) particle displacements in a trajectory. (For exclusion processes, this is the difference between the numbers of right and left hops.) For generality, we consider jointly biased ensembles where the activity is biased by a field s and the current J is biased by a field h. The analogue of (1) is $\langle O\rangle_{s,h} = e^{-\psi_{KJ}L^d t_{\mathrm{obs}}}\langle Oe^{hJ-sK}\rangle_0$ [22]. Within fluctuating hydrodynamics (assuming $a = 0$ as above), the response to the bias depends only on the quantity $B = s\kappa_0'' - \frac{1}{2}h^2\sigma_0''$ [22]. For $B = 0$, then $S(q)$ has a finite (nonzero) limit as $q \to 0$; for $B > 0$, the system is hyperuniform, while for $B < 0$, one has phase separation. The resulting dynamical phase diagram is shown in Fig. 2: the fluctuating

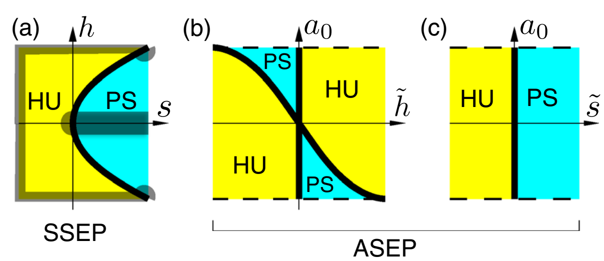

FIG. 2 (color online). Proposed dynamic phase diagrams for biased exclusion processes. HU, hyperuniform states; PS, phase separation. (a) SSEP, jointly biased by both activity and current. On the heavy black line, the system has normal fluctuations. In the shaded regions, the indicated behavior can be shown analytically. (b) ASEP with hopping asymmetry a_0, biased by the current. Normal fluctuations occur for zero bias ($\tilde{h} = 0$) and on the line $a_0 = -\tanh(\tilde{h}/2)$, which is related to $\tilde{h} = 0$ by the Gallavotti-Cohen symmetry. (c) ASEP, biased by the activity. Normal fluctuations occur only for zero bias.

hydrodynamic analysis holds only for $s, h \ll 1$, but in the absence of additional phase transitions, one expects the same structure to hold throughout the (s, h) plane. We discuss this conjecture below, using results from exclusion processes.

The condition $B = 0$ recovers a homogeneous state with $S(q \to 0) > 0$: we use the term "normal fluctuations" for this case, in contrast to hyperuniform or phase-separated states. In fact, biased ensembles with $B = 0$ are identical to (unbiased) steady states of models in which time-reversal symmetry is broken ($a \neq 0$). This may be verified directly from (3), but a clearer interpretation of this result can be obtained by analyzing exclusion processes, as we now discuss.

Mappings between biased ensembles for exclusion processes.—We analyze exclusion processes via operator representations of their master equations [40]. Starting from the master equation for the SSEP, we write a biased generator [5,27] that describes the jointly biased ensemble. To account for the bias that appears in (1), this operator must correspond to a time evolution that does not in general conserve probability [5,27]. It has a representation in terms of Pauli spin matrices [22]:

$$W_S(s,h) = \sum_i e^{h-s}\sigma_i^-\sigma_{i+1}^+ + e^{-h-s}\sigma_i^-\sigma_{i-1}^+ - 2n_i(1-n_{i+1}), \quad (6)$$

where $n_i = \sigma_i^+\sigma_i^-$. If we consider instead an ASEP biased by its current, the relevant operator is $W_A(\ell, r, \tilde{h}) = \sum_i r e^{\tilde{h}}\sigma_i^-\sigma_{i+1}^+ + \ell e^{-\tilde{h}}\sigma_i^-\sigma_{i-1}^+ - (r+\ell)n_i(1-n_{i+1})$, where ℓ, r are hopping rates and $\tilde{h}$ the biasing field. Note the Gallavotti-Cohen symmetry [5,27,41]: $W_A(\ell, r, \tilde{h}) = W_A[r, \ell, \tilde{h} - \log(r/\ell)]$. For appropriate parameters, we may have $W_S = W_A$, which means that the trajectories of the two biased ensembles are identical. Defining the hopping asymmetry $a_0 = (r-\ell)/(r+\ell)$, equality between W_S and W_A requires $a_0 = \tanh(h - \tilde{h})$ and $e^s = \cosh(h - \tilde{h})$. Any jointly biased SSEP with $s > 0$ leads to two solutions for $\tilde{h}$, which correspond to two possible current-biased ASEPs.

The SSEP with $\cosh h = e^s$ is a special case because it corresponds to an unbiased ASEP ($\tilde{h} = 0$). This mapping provides a microscopic interpretation of the condition $B = 0$ in the fluctuating hydrodynamic analysis [for small h, s, we obtain $s = h^2/2$, which is consistent with $B = 0$, because $\sigma(\rho) = \kappa(\rho)$ for the SSEP]. The unbiased steady state of the ASEP has normal fluctuations, so we conclude that fluctuations in the jointly biased SSEP are also normal if $\cosh h = e^s$ [solid line in Fig. 2(a)].

There is a family of mappings between biased exclusion processes [22]: any activity-biased ASEP may also be mapped to a jointly biased SSEP. The resulting situation is shown in Figs. 2(b) and 2(c) where we show the ASEP dynamical phase diagrams that correspond to the (conjectured) phase diagram in Fig. 2(a). The hypothesis is that all points in Fig. 2(a) are either phase separated or hyperuniform, except for the normal line $\cosh(h) = e^s$. For these purposes, phase-separated states are those with macroscopically inhomogeneous density profiles, which might include stationary clusters of particles, or "traveling-wave" states, where a large cluster moves through the system with finite velocity [42,43]. The fluctuating hydrodynamic analysis establishes these results in the small bias regime $|h|, |s| \ll 1$, as discussed above. The question is therefore whether some other phase transition might intervene for $h, s = O(1)$, destroying the phase-separated or hyperuniform states.

We are not able to rule out this possibility, but several exact results indicate strongly that there is no such phase transition. (i) For $s \to -\infty$, the density correlations of the ASEP are known [44]; independently of the asymmetry a_0, there is a logarithmic effective interaction potential between particles which renders this state hyperuniform. This implies that the jointly biased SSEP is hyperuniform as $s \to -\infty$ (for all h), and the same analysis also holds for $|h| \to \infty$ at fixed s. (ii) For $h = 0$, a variational argument [4] indicates that the SSEP phase separates for all $s > 0$ because the system can then access configurations where the total number of available hops remains finite as $L \to \infty$. (iii) Phase separation has been shown analytically for the totally asymmetric exclusion process ($a_0 = 1$); this transition corresponds to the appearance of "shocks" in response to a bias on the current [42]. For the SSEP, this establishes phase separation for all $B > 0$ in the limit $h \to \infty$. Combining these results establishes that the proposed phase diagram of Fig. 2(a) is correct in all of the shaded regions: we cannot rule out other phase diagrams that are consistent with these constraints, but this simple picture is the most likely scenario. If the proposed Fig. 2(a) is correct, the phase diagrams in Figs. 2(b) and 2(c) follow from the exact mappings between biased exclusion processes.

Conclusion.—Figure 2 indicates that exclusion processes respond very strongly to biases h and s, which almost always lead to either phase-separated or hyperuniform states. The normal fluctuations that are familiar from equilibrium systems occur only under special high-symmetry conditions, such as $B = 0$. These results provide another example [16,19,20] of hyperuniformity emerging in nonequilibrium states, and they show that the dynamical phase transition identified in Ref. [21] corresponds to the appearance of hyperuniformity. More generally, the theory of fluctuating hydrodynamics indicates that these dynamical phase transitions should be generic ("universal") in systems with locally conserved hydrodynamic variables such as energy or density—all results presented here are for systems with periodic boundary conditions, but some similar effects also occur in the ASEP with open boundaries [45]. The interplay between these phase transitions and the "glass transitions" found previously in biased ensembles of trajectories [4,7,37] merits further study—diffusive large-scale behavior is not a necessary condition for those glass transitions [4,46], but the analysis presented here indicates that phase-separated states may compete with homogeneous glassy states in systems that are biased to low activity.

We thank Fred van Wijland, Vivien Lecomte, and Juan P. Garrahan for many useful discussions. R. L. J. and I. R. T. were supported by the EPSRC through Grant No. EP/I003797/1.

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