

Universality Class of the Reversible-Irreversible Transition in Sheared Suspensions

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Collections of non-Brownian particles suspended in a viscous fluid and subjected to oscillatory shear at very low Reynolds number have recently been shown to exhibit a remarkable dynamical phase transition separating reversible from irreversible behaviour as the strain amplitude or volume fraction are increased. We present a simple model for this phenomenon, based on which we argue that this transition lies in the universality class of the conserved DP models. This leads to predictions for the scaling behaviour of a large number of experimental observables. Non-Brownian suspensions under oscillatory shear may thus constitute the first experimental realization of an inactive-active phase transition which is not in the universality class of conventional directed percolation.

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The equations of fluid dynamics become time-reversible in the Stokesian limit where dissipation dominates and inertia is ignored [1]. Neutrally buoyant non-Brownian particles in a sheared Stokesian fluid, do, nonetheless, diffuse [2, 3, 4, 5]. This irreversible behaviour is the result of an infinite sensitivity [6] to initial conditions.

In ref. [7] a suspension of neutrally buoyant PMMA spheres (diameter 200 μm , Brownian diffusivity $\sim 10^{-14}$ cm^2/s) in a Newtonian fluid of viscosity 3 Pa s, was subjected to periodic shear $\gamma(t) = \gamma \sin \omega t$. The Reynolds number in the experiments was less than 10^{-3} , justifying the Stokesian approximation. At low volume fraction ϕ or γ the trajectories were observed [7] to be reversible, but at large enough ϕ or γ they were seen to be chaotic, irreversible, and consistent with diffusive behaviour. These two regimes were found to be separated by a remarkable continuous nonequilibrium phase transition. More recently Corté *et al.* showed that some initial random motion always arises upon application of oscillatory shear. However, for small enough ϕ and γ this motion is transient, disappearing with a relaxation time τ that diverges as a power law as the transition is approached. For large enough ϕ and γ a nonzero level of activity persists indefinitely. The universality class of this unique dynamical phase transition is the subject of this paper.

The physics underlying this transition is well described by an elegant simulational model [8] of point particles in a square domain subjected to sinusoidal shear, and suffering random displacements if they come within a specified distance of each other. For small γ and ϕ , most particles

never meet another, and passively follow the imposed oscillatory strain. Those few particles which happened initially to be close together will undergo encounters and move apart, and the particle positions will settle down into one of a potentially infinite number of inactive or “absorbing” configurations. In such configurations, all particles are far enough apart that their relative positions are not disrupted over a cycle of periodic strain. As γ or ϕ is increased, absorbing configurations become rarer, so the time to settle into a quiescent state grows. Across a threshold strain amplitude $\gamma_c(\phi)$, the effect of close encounters propagates throughout the system, stimulating persistent, global particle diffusion.

The reversible-irreversible behaviour seen in the experiment is thus explicitly a transition from a highly degenerate absorbing state to an active state [8]. (An individual particle is said to be active if it has moved perceptibly as a consequence of its interaction with other particles as the suspension is sheared through a full cycle.) The directed percolation (DP) transition [9, 10, 11] was conjectured to be the generic transition out of a highly degenerate absorbing state. Systems like those of [7, 8] in which activity is carried by the motion of a locally conserved quantity such as particle number, however, are expected to lie in a universality class distinct from DP [12, 13]. The key proposal of this paper is that the transition seen in the experiments should lie in the universality class of such *conserved* DP models, the CDP class.

We motivate our model by abstracting what we believe are the essential details of [7, 8]. The experiments sample density configurations stroboscopically, at time intervals fixed by the periodicity of the imposed strain, thus focusing only on that component of individual particle motion which is determined by its interactions with the other particles. A particle which does not have other particles in its close vicinity executes reversible motion over the time period of the imposed strain; measured stroboscopically, the particle is at rest. In regions of larger volume

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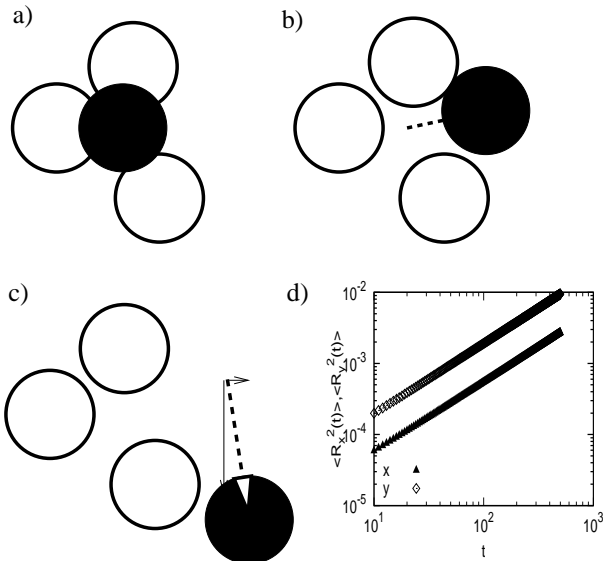


FIG. 1: Update of particles in the off-lattice, anisotropic model for conserved directed percolation. A particle (black) with one or more other particles in its interaction radius R_0 (Fig. 1(a)) is updated by first giving it a random displacement in a direction chosen so as to correspond to a local decrease of the density (Fig. 1(b)). The particle is also further displaced by a random vector, whose magnitude is chosen from a uniform zero-mean distribution with different widths in x and y directions, as shown in Fig. 1(c). Fig. 1(d) illustrates the anisotropic diffusive behaviour of particles above the active-inactive transition where the mean-square displacement in the x - and y - direction, averaged over all particles, as $R_x^2 = \frac{1}{N} \sum_i \langle [R_x^i(t) - R_x^i(0)]^2 \rangle$. The anisotropy $D_y/D_x = 3.8$ in the data shown, comparable to the values obtained in the experiments.

fraction, particle displacements in close encounters are in effect random.

A simple model follows from the physical picture described above. Consider a lattice with sites which can be occupied either by one particle or none (a vacancy). Particles with no nearest neighbour site occupied do not move. Particles occupying adjacent sites are moved to randomly chosen empty sites which neighbour them. A particle is tagged as active if it has moved in the previous time step. All sites are updated in parallel. The fraction of occupied sites, which we shall call the concentration, is the sole control parameter. The question of interest is: For generic initial conditions, is there residual activity in the particles as $t \rightarrow \infty$?

The model defined above has been studied earlier [15, 16, 17]. As the concentration is increased, the model exhibits an inactive-to-active phase transition which is *not* in the DP universality class [16, 17, 18, 19]. This is because the activity, not a conserved quantity, is carried by particles whose number is locally conserved.

The lattice model can be mapped to a continuum

stochastic dynamical field theory [13, 20]:

$$\begin{aligned} \frac{\partial A}{\partial t} &= D^A \nabla^2 A + \mu A - \lambda A^2 + \kappa \rho A + \sigma \sqrt{A} \eta \\ \frac{\partial \rho}{\partial t} &= D^\rho \nabla^2 A \end{aligned} \quad (1)$$

where $A(\mathbf{r}, t)$ denotes the activity field and $\rho(\mathbf{r}, t)$ the local number density of particles. The quantities $D^A, D^\rho, \mu, \lambda$ and κ are constants – in general depending on concentration – and $\eta(\mathbf{r}, t)$ is a spatiotemporally white Gaussian noise. These equations encode the following: (i) particle motion carries activity to nearby particles; (ii) local density promotes local activity through κ ; (iii) activity gradients produce particle diffusion; and (iv) noise arises only in regions with activity and, hence, with particles. Numerical integration of the equations yields exponents which coincide with the results from the several versions of the lattice models which have been proposed to exhibit conserved DP scaling[21].

The average activity $\langle A \rangle$ is defined in the lattice model [13] as the time-averaged fraction of active particles, and in the continuum model [21] as the steady-state mean of the field A . It is found that $\langle A \rangle$ is zero below the transition and non-zero above it, with a continuous but non-analytic onset: $\langle A \rangle \sim (\rho - \rho_c)^\beta$ where β is a universal critical exponent. Close to this transition, fluctuations in the activity are correlated over distances $\xi \sim (\rho - \rho_c)^{-\nu}$, and times $\tau \sim \xi^z$. Simulations of the lattice models and the equivalent continuum equations given above find $\beta \simeq 0.84$ in dimension $d = 3$ and $\beta \simeq 0.64$ in $d = 2$ [13]. The correlation length exponent $\nu \simeq 0.59$ ($d = 3$) and 0.79 ($d = 2$) while the dynamical exponent $z \simeq 1.82$ ($d = 3$) and 1.53 ($d = 2$) [13]. The mean-field values for both DP and CDP models are $\beta = 1, \nu = 1/2, z = 2$.

How well is the physics of the experiments of [7, 8] incorporated in the equations above? It is clear that the detailed trajectories, governed by the Stokesian dynamics of particles and fluid, are irrelevant to understanding the transition itself. All that matters is whether the particles return to their original states upon stroboscopic sampling at the frequency of the applied strain, or have moved randomly as a consequence of interactions during the strain cycle. The essence of this physics is adequately included in the lattice model and its field theoretic translation. It is important that the physics of the transfer of activity is local – in the experiments, activity initiated in a local region of the sample only affects contiguous regions. Since the experiments are performed in a Couette geometry with a narrow gap w in the gradient direction, the hydrodynamic interaction is highly attenuated on scales larger than w in the remaining two (axial and circumferential) directions. We shall comment below on what to expect when all sample dimensions are comparable.

Mapping the experiment to the model requires two key assumptions. First is the reasonable expectation that driving such a highly overdamped system at nonzero frequency should be irrelevant to the hydrodynamic behaviour once averaged over the driving period [22]. Sec-

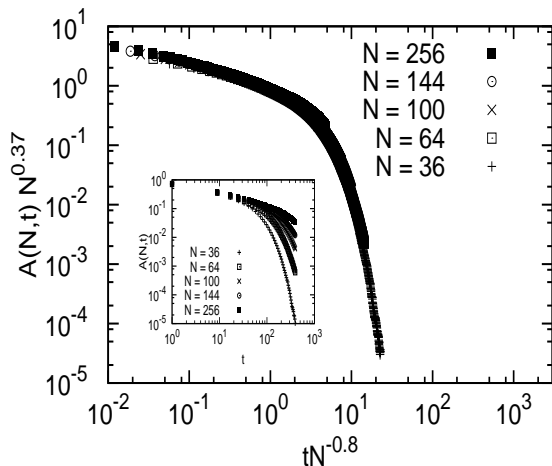


FIG. 2: Scaling collapse of the fraction of active particles $A(N, t)$ for particle numbers $N = 36, 64, 100, 144$ and 256 , plotted as $A(N, t)N^{\alpha z/2}$ vs. $tN^{-z/2}$. The data collapse illustrated is obtained for $\alpha = 0.42$ and $z = 1.6$. The inset shows the bare data corresponding to the scaling plot.

ond and perhaps more relevant is our assumption of isotropic diffusion. The motion induced by close encounters between particles is not isotropic even in the velocity-vorticity plane. Is such anisotropy relevant at asymptotically large length- and time-scales? This reduces to the question of whether the anisotropies of the diffusivities for activity and density are equal at such scales.

We examine these issues through simulations of an off-lattice model of conserved DP which mimics our stroboscopic interpretation of the simulation model of Corté *et al.* Our model considers N particles in a square simulation box of area \mathcal{A} , each with an interaction radius R_0 , thus defining a coverage NR_0^2/\mathcal{A} . Every particle with one or more other particles in its interaction radius (Fig. 1(a)) is updated in parallel as shown in Figs. 1(b) and (c), by giving it a random displacement in a direction chosen so as to correspond to a local decrease of the density (Fig. 1(b)). In addition, the particle is also further displaced by a random vector, whose magnitude is chosen from independent uniform distributions in x and y with widths W_x and W_y which are, in general, unequal. As a consequence, above the threshold, particle motion is anisotropic and diffusive, as illustrated in Fig. 1(d), where the mean-square displacement in the x - and y -direction, averaged over all particles is shown separately.

The scaling behaviour of our off-lattice, anisotropic generalization of the CDP model can be studied through the scaling ansatz, valid at the critical point

$$A(N, t)N^{\alpha z/2} = \tilde{F}(a_N t N^{-z/2}) \quad (2)$$

where the factors of $z/2$ follow from the fact that we work at fixed number of particles N , varying R_0 so as to maintain the coverage. The inset of Fig. 2 shows the

time-dependence of the fraction $A(N, t)$ of active particles as a function of time at the critical point. This data, obtained by averaging over $10^6 - 10^7$ independent runs, is plotted using the scaling ansatz of Eq. 2 in the main panel of Fig. 2, for an anisotropy $D_y/D_x \sim 3.5$. Data collapse is obtained for $\alpha = 0.42 \pm 0.02$ and $z = 1.6 \pm 0.1$, to be compared to the best values obtained from lattice simulations of $\alpha = 0.423$ and $z = 1.54$. Scaling fits for smaller values of the anisotropy $D_y/D_x \sim 1.6$, yield exponent values $\alpha = 0.42 \pm 0.01$ and $z = 1.55 \pm 0.05$; for zero anisotropy values within the same range are obtained. Thus, we conclude that anisotropies comparable to those in the experiments do not affect the critical exponents of the isotropic CDP transition and that our anisotropic off-lattice CDP model is in the same universality class as the lattice model.

The experimental system, in principle, allows us to explore the full range of dimensionalities and dimensional crossovers in the conserved DP problem, through the reduction of one or more dimensions of the Couette cell in which the experiments are done. If the gap thickness w and radius R of the cylinder are kept small and fixed and the length $L \gg w, R$, the exponents that emerge should be compared to those of the one-dimensional C-DP model. If instead $R \sim L \gg w$, the system is effectively two-dimensional. The experiments of Corté *et al.* are performed in a quasi-two-dimensional geometry in which the gap between the two cylinders in the Couette cell is the smallest relevant length scale, corresponding to about 11 particle diameters. The growth of relaxation times close to the active-inactive transition should follow from the scaling exponents outlined above, with $\tau \sim (\rho - \rho_c)^{-1.22}$ for $d = 2$. For this exponent, Corté *et al.*, find a value close to 1.2, in encouraging agreement with our predictions.

Experimentally, Corté *et al.* find the order-parameter exponent β to be close to the mean-field prediction of 1, but this estimate depends on a threshold criterion for the presence of particle motion. In addition, the experimental transition is probably rounded for reasons we discuss below. The simulations of the two-dimensional model proposed by Corté *et al.* obtain an exponent of $\beta \simeq 0.45$, probably without a detailed finite-size scaling analysis; recent related work [29] finds 0.59, closer to our predicted value. The mapping to CDP provides predictions for several other possible experiments involving the evolution of activity under local perturbations in systems at the critical point: the average number of active sites $N(t) \sim t^{-\theta}$ and the survival probability $P(t) \sim t^{-\delta}$ of the activity after time t . Here the exponent values are $\theta \simeq 0.31$ ($d=2$) and $\theta \simeq 0.14$ ($d=3$) while $\delta \simeq 0.51$ ($d=2$) and $\delta \simeq 0.76$ ($d=3$)[13]. Experiments which involve examining the behavior of the activity under such local perturbations about inactive states do appear to be possible[23], and would be an important test of the ideas proposed here.

Some important caveats: In a Couette cell with gap w , the hydrodynamic interaction between particles does not

vanish abruptly at a finite distance but is merely reduced substantially on scales larger than w in the remaining directions [25, 26]. Small perturbations in particle positions may then be communicated weakly over infinite distances. A tiny irreversibility could thus persist [24] at the smallest values of γ and ϕ , and its cumulative effect over many strain cycles should be observable, as weak diffusion of the density in the nominally inactive regime. Such an additional diffusion is analogous to a weak field conjugate to the order parameter in conventional critical phenomena, turning the sharp transition into a rapid crossover [27]. The experiments of [7, 8] may well be seeing this *rounded* transition from weak to strong irreversibility [28]. The nonequilibrium phase boundary reported in [7, 8] would then simply be the locus of points in the (γ, ϕ) plane below which the time taken for irreversible behaviour to manifest itself exceeds the time-scale of the experiment.

The effectively one-dimensional case $L \gg w, R$, where hydrodynamic screening is expected [25] to be exponential, should show behaviour closest to a genuine transition. The long-ranged nature of the hydrodynamic interaction will manifest itself fully in the “three-dimensional” limit $L \sim R \sim w \rightarrow \infty$; we cannot be sure that the 3-dimensional C-DP model with local interactions applies to this case. We note that the Stokesian simulations of PGBL, performed using periodic boundary conditions and thus in effect simulating the three-dimensional system, show Lyapunov exponents which, though small, are still non-zero even at the lowest shear amplitudes. Ex-

periments and simulations which probe the variation of the critical shear rate for the onset of the transition, with w, R and L varied so as to interpolate between one and three-dimensional behaviour should illuminate the role played by the far-field part of the hydrodynamic interaction and the role of effective dimensionality in this problem.

In conclusion, we have suggested that the universal behaviour which should underly the experiments and simulations of [7, 8] is to be identified with the universality class of conserved directed percolation [13]. This identification leads to specific predictions for the exponents of the active to inactive transition in the experiments. We thus propose that these experiments constitute the first experimental realization of a system in the universality class of C-DP.

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