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Irreversibility and self-organization in hydrodynamic echo experiments

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We discuss the reversible-irreversible transition in low-Reynolds hydrodynamic systems driven by external cycling actuation. We introduce a set of models with no auto-organization, and show that a sharp crossover is obtained between a Lyapunov regime in which any noise source, such as thermal noise, is amplified exponentially, and a diffusive regime where this no longer holds. In the latter regime, groups of particles are seen to move cooperatively, yet no spatial organization occurs.

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The behavior of a system that retraces its steps after a reversal in its dynamics—the echo protocol—has been of long-standing interest. From a fundamental perspective, the question of how macroscopic irreversibility arises from the reversible dynamics of the microscopic components led to one of the central debates on the foundations of statistical mechanics between Loschmidt and Boltzmann [1]. Loschmidt argued that reversing the velocities of all the particles in a box should allow for the system to return to its initial position, thus invalidating the notion of an arrow of time. In later years, echo experiments became practical tools as nuclear magnetic resonance and neutron spin-echo techniques to probe irreversible microscopic events in soft materials, and has also, as the name would suggest, numerous applications in acoustics [2]. From a numerical perspective, Levesque and Verlet showed how rounding off errors suffice to destroy reversibility in classical Hamiltonian systems [3]. A conceptually closely related situation, in the field of hydrodynamics, is the classic Taylor experiment to demonstrate the perfect reversibility of viscous fluid flows [4]. It consists of adding a drop of dye to a viscous fluid in the gap between two concentric cylinders. The drop is then strongly stretched by turning the inner cylinder. By subsequently imposing the reverse rotation, mixing is not enhanced: on the contrary, the initial spherical shape of the drop is recovered. A rather spectacular elaboration of this experiment was recently performed by Pine *et al.* [5], who substituted the ink droplet with a high volume fraction of non-Brownian beads. Although shear flow induces an effective diffusion of the particles in the suspension due to hydrodynamic interactions [6,7], the complicated particle trajectories thus generated during half a cycle should in principle be retraced in the second half. By measuring the net particle displacement after each back and forth cycle, Pine *et al.* uncovered a remarkable transition from a reversible situation in which particles do retrace their steps to a regime obtained above a critical strain amplitude, in which reversibility is lost.

Two different but related questions immediately arise: (i) what is the origin of irreversibility, and (ii) is the transition a sharp one, and if so, what is its nature? In a recent paper, Corte *et al.* proposed that the key point could be the close particle encounters, where nonhydrodynamic—and hence nonreversible—contact interactions (Van der Waals, mechanical friction) act [8]. In one stroke this provides both an explanation for irreversibility and a mechanism for a sharp transition: because particles move away from situations that generate diffusion, they self-organize in subsequent cycles into a configuration in which they avoid encounters, and once this is done irreversible diffusion stops. Above a certain percolation-like transition, the system is no longer able to find such a configuration without collisions, and irreversibility persists in time. This scenario was further studied analytically by Menon and Ramsawami [9], along the lines of [10].

In this Rapid Communication, we test the opposite scenario: we study models that, by construction, cannot lead to self-organization. We assume that small irreversible perturbations, such as Brownian motion, exist all along and are amplified by chaoticity. This will take us to a situation that is very close to that of echoes in classical Hamiltonian systems. We shall show that a sharp increase of particle diffusion can be observed in such a "hydrodynamic echo experiment" even if no real dynamic phase transition and no self-organization process occur. Although it is likely that the contact-induced self-organization scenario is relevant for the experiments in Ref. [5], we believe that other experimental situations (and perhaps even the numerical hydrodynamic simulation in [5]) may well correspond to simple chaotic amplification.

Consider N force-free particles immersed in a viscous incompressible fluid bounded between two concentric cylinders, the inner one being able to rotate at frequency $\omega(t)$. For spherical particles and creeping flows, the particle velocities are linearly related to ω [11],

$$\dot{\mathbf{x}}_a = M_a(\mathbf{x}_1, \ldots, \mathbf{x}_N)\omega, \tag{1}$$

where the $\mathbf{x}_a$ and $\dot{\mathbf{x}}_a$ are the particle center positions and velocities. We shall henceforth set $|\omega|$ to 1. The mobility coefficients M_a are complex vectorial functions, obtained in principle by solving the stationary Stokes problem with the instantaneous boundary conditions, and eliminating the angular velocities using the fact that the particles are torque-free. The evolution is reversible if we make half a period $T/2$ with ω and subsequently $T/2$ with $-\omega$; each particle retraces its steps over a cycle as in Taylor's experiment. Next, consider the effect of a noise $\eta_a(t)$ of small amplitude, ϵ, acting

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on the particle a that we shall assume white but in general with spatial correlation $\langle \eta_a(t)\eta_b(t')\rangle = 2\epsilon^2\delta(t-t')\tilde{M}_{ab}(\mathbf{x}_a,\mathbf{x}_b)$. In the specific case of the thermal noise, $\tilde{M}_{ab}(\mathbf{x}_a,\mathbf{x}_b)$ is equal to the mobility matrix, such as to respect the fluctuation-dissipation theorem [12]. The equation of motion for the N particles is

$$\dot{\mathbf{x}}_a = \begin{cases} +M_a(\mathbf{x}_1,\ldots,\mathbf{x}_N) + \eta_a(t) & \text{if } 0 < t < T/2, \\ -M_a(\mathbf{x}_1,\ldots,\mathbf{x}_N) + \eta_a(t) & \text{if } T/2 < t < T. \end{cases} \tag{2}$$

For $\epsilon \neq 0$, given that the noise is different in the two semicycles, the trajectory does not retrace its steps exactly. How much does a trajectory deviate from the initial position after a cycle? Importantly, the invariance upon time reversal of Eq. (2) tells us that this question is equivalent to asking how much two trajectories, starting from the same position at midcycle, diverge under the effect of different noise realizations. Let us first make this discussion for small noise and for large cycle times. In this limit, we can express the separation, Δ, of the particles between the two trajectories in the $3N$-dimensional phase space in terms of the Lyapunov exponents [13],

$$\Delta \propto e^{\lambda^{\text{traj}}T/2}, \tag{3}$$

where λ^{traj} is the largest Lyapunov exponent. An important remark that should be made is that, because λ^{traj} is a function of the trajectory, depending on the specifics of the driving, it may or may not coincide with the one of a randomly chosen trajectory after a few cycles. For instance, the system may drift away from the regions with high Lyapunov exponents and converge to a subset of the phase space corresponding to smaller diffusivity. We will refer to such a phenomenon as *self-organization*.

We now restrain ourselves to a limiting case in which self-organization does not happen. Let us consider the limit of pointwise particles, or equivalently of very dilute suspensions. In this limit, the particles behave like fluid tracers. It thus follows from the (fluid) incompressibility conditions that the mobility coefficients obey

$$\sum_a \nabla_a M_a = 0, \tag{4}$$

with $\nabla_a \cdot \equiv \partial\cdot/\partial_{\mathbf{x}_a}$. This implies that the probability distribution $P(\mathbf{x}_1,\ldots,\mathbf{x}_N)$ converges to the flat measure. Indeed, during each half-cycle, P evolves according to the Fokker-Planck equation [12],

$$\dot{P} = \sum_{ab} \nabla_b\{\epsilon^2\tilde{M}_{ba}\nabla_a \pm \delta_{ab}M_a\}P, \tag{5}$$

which admits a constant function as a solution by virtue of Eq. (4). In this case, there can be no self-organization. In fact, if the initial positions are chosen with flat probability, the probability distribution does not evolve and no *static* one-time correlation function depends on the cycle number. Note that there are two independent issues concerning external forces and interparticle potential interactions: (i) whether they break time reversal, and (ii) whether they preserve the flat measure. Needless to say, in a system with divergenceless forces there can be no structural phase transitions other than those of equilibrium hard spheres. Furthermore, the separation of nearby trajectories corresponds to that of an equilibrium unbiased system, and in particular Lyapunov exponents are the "typical" ones sampled in equilibrium. Because of the obvious analogy with the well-studied classical and quantum-mechanical problem, we shall call this the hydrodynamic Loschmidt-echo situation.

To stress the analogy, in this Rapid Communication we shall substitute hydrodynamic evolution, Eq. (2), by simple Hamiltonian dynamics, in the presence of random noise, such that it conserves the microcanonical distribution. Our first example is very similar to the original Loschmidt experiment: we consider particles in two dimensions with power law r^{-3} interaction, perturbed by small energy-conserving noise (the choice of a long-range interaction, by analogy with hydrodynamic coupling, is intended to limit the role of particle encounters). The second example, which allows us to go to large sizes and times, is a system of coupled simplectic maps [14,15].

Consider first the two-dimensional (2D) system. We perform direct followed by time-reversed evolution, after a velocity reversal in midcycle. If the system is started in a thermalized microcanonical configuration, then configurations are statistically distributed in the same way at all times. Figure 1 shows the average quadratic displacement in one cycle in terms of the cycle time, for two values of noise amplitude. The shape of the curve is very similar to the one reported for sheared suspensions and for superconducting vortices [5,16]. The mean squared displacement first increases exponentially with T and then saturates to a constant value.

This behavior can be explained as follows. At short periods, back and forth trajectories differ only slightly, and the Lyapunov linearization applies. Moreover, the separation per particle is stable in the thermodynamic limit, everything else (period, particle, and energy density) being kept equal. Indeed, it can be shown that the condition for this is that the distribution of the Lyapunov exponents converges to a stable "Lyapunov density function," which is expected for a many-particle system [13,17].

At times T^* such that $\epsilon e^{\lambda T^*}$ is a sizable fraction of the interparticle distance, the linearization breaks down [18]. Moreover, for $T \gg T^*$ the system completely loses memory in a cycle, and the separation becomes "diffusive" rather than exponential. Note, first, that the apparent transition time $T^* \sim \lambda^{-1}\ln(1/\epsilon)$ depends very weakly on the irreversible noise amplitude, and, second, that it has a well-defined (large-N) thermodynamic limit. In turn, the main features of the fluctuations of the stroboscopic positions with T are insensitive to the specific process that breaks the time-reversal symmetry. Note also that the saturation of the actual particle *self*-diffusion occurs with the typical displacement of order of the box size. If we now make plots with the y axis in linear scale and normalized by the saturation, the curve will have a sharp inflection; see Fig. 1. In this sense, and probably only in this sense, there is a sharp transition without self-organization.

In Fig. 2, we show the same results for a toy model: a time-discrete Hamiltonian system of N coupled maps constructed by the composition of the following two one-step iterations and their time reversal [15]:

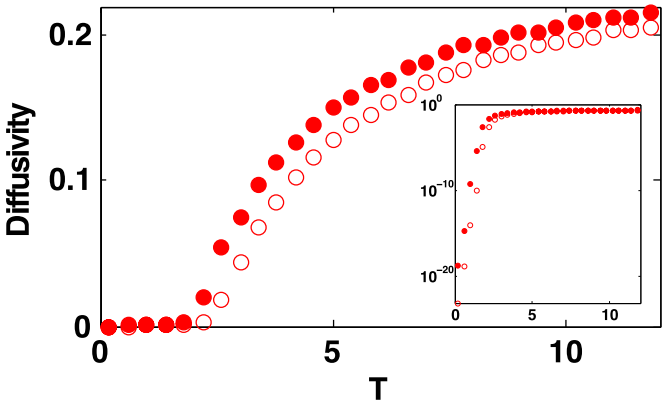

FIG. 1. (Color online) Diffusivity D vs period T for the 2D system composed of $N=250$ particles. Over each half-cycle, the particle positions evolve according to $\ddot{\mathbf{x}}_a=-\Sigma_b\alpha|\mathbf{x}_b-\mathbf{x}_a|^{-4}+\epsilon_a$, where the ϵ_a are uncorrelated Gaussian white noises of variance ϵ and $\alpha=2\times10^{-6}$. The box size is set to 1 and the energy is chosen equal to $E\approx87$. The diffusivity is averaged over 100 cycles. The open symbols correspond to the zero additional noise limit, $\epsilon=0$, yet the trajectories are not reversible. This is due to the (irreversible) numerical rounding off [3]. The filled symbols correspond to $\epsilon=(1/\sqrt{2})\times10^{-7}$. Inset: same plot in log scale.

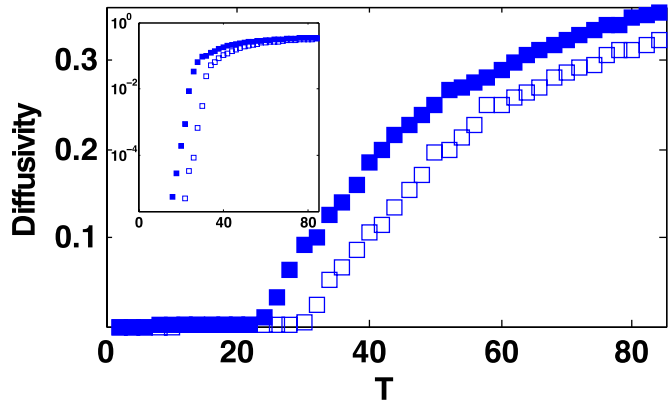

FIG. 2. (Color online) Diffusivity D vs period T for $N=7000$ coupled maps. The diffusivity has been averaged over 500 cycles. Open (filled) symbols correspond to a noise amplitude $\epsilon=(1/\sqrt{2})\times10^{-12}$ [$\epsilon=(1\sqrt{2})\times10^{-10}$]. Inset: same plot in log scale.

$$\left.\begin{aligned} p_i' &= p_i + \epsilon\xi_{p_i} \\ q_i' &= q_i + p_i + \epsilon\xi_{q_i} \end{aligned}\right\}\text{mod } 1, \tag{6}$$

$$\left.\begin{aligned} p_i' &= p_i' + \frac{1}{\sqrt{N-1}}\sum_{j=1}^{N}\sin[2\pi(q_i'-q_j')] \\ q_i'' &= q_i' \end{aligned}\right\}\text{mod } 1, \tag{7}$$

where $q_i'(t)\equiv q_i(t+1)$, $p_i'(t)\equiv p_i(t+1)$. ξ_{p_i} and ξ_{q_i} are Gaussian white noise of variance 1. Again the fluctuations of the stroboscopic position in the (q,p) plane display the same features. Note that for this simple model we achieved $N=7000$ and have checked that the curves for different (large) N coincide within numerical error, and we did not observe any sharpening of the crossover. Figure 3 shows the motion of particles just above the mobility time. Surprisingly enough, even in the absence of self-organization, the particle system seems not entirely devoid of spatial features. At a given time, there are three types of particles: almost immobile, those that move *back and forth* along essentially one-dimensional trajectories, and those that have a more typical diffusive character. This latter group—which becomes dominant well above the crossover—looks spatially correlated, suggesting that these are the particles that interact the most along trajectories. In Fig. 4, we show the probability distribution function of the individual particle diffusivity in one cycle. The distribution seems exponential, and there are relatively many highly diffusive particles at any time. The observation of some degree of dynamic heterogeneity does not contradict the fact that static correlations are those of the equilibrium system, since this involves a purely dynamic correlation, the spatial correlations between particle *displacements* in a cycle.

The scenario discussed here is by construction the opposite of the one considered in Refs. [8,9], which involves a dramatic change in the stationary end-of-cycle distribution near the transition, completely absent here. In Ref. [8], the time for reaching the stationary value of diffusivity was plotted, showing a peak near the transition strain, an evidence of organization. The situation could, however, be mixed: consider a system with perfect time-reversal broken by a constant Brownian noise, so that the flat distribution is preserved. Suppose we switch on forces that violate the uniform distribution: this is indeed what happens in the hydrodynamic case as the volume fraction is increased. The system will now self-organize into a distribution that, unlike the case we studied here, will depend on strain, noise level, and particle volume fraction. If the effect is weak, we do not expect that a sharp transition will immediately arise, so that from this point of view the scenario is still the one we discussed here, in spite of having a certain degree of auto-organization. In order to further emphasize the experimental relevance of the hydrodynamic echo scenario, we compute the crossover shear amplitude γ^*, which would correspond to the experimental conditions reported in [5,8]. The noise source is supposed to be the weak Brownian motion of the suspended particles. ϵ is thus approximated by the distance over which a single particle diffuses over half a cycle: $\epsilon\equiv\sqrt{DT}$, $D=k_BT/(6\pi\eta a)$ with $\eta=3$ Pa s the fluid viscosity and $a=115$ μm the particle mean radius [8] (note that we have neglected hydrodynamic correlations of the noise). The interparticle distance is of order of $\Delta\sim a/\phi^{1/3}$. To compute γ^*, we also need the Lyapunov exponent of the concentrated particle suspension. This value has been computed numerically in [19] for $\phi=0.4$: $\lambda\sim0.6\dot{\gamma}$, where $\dot{\gamma}\equiv2\pi\gamma/T$ for sinusoidal cyclic shear. We thus infer a critical shear amplitude of $\gamma^*\sim3$, which is fairly close to the experimental value $\gamma^*_{\text{exp}}\sim1$ given the crudeness of our numerical estimates. These predictions suggest that there are very probably many situations intermediate between the percolation-like transition and the pure Loschmidt echo crossover discussed in this Rapid Communication, the latter being itself not far from the situation of a single particle advected in a chaotic time-reversed

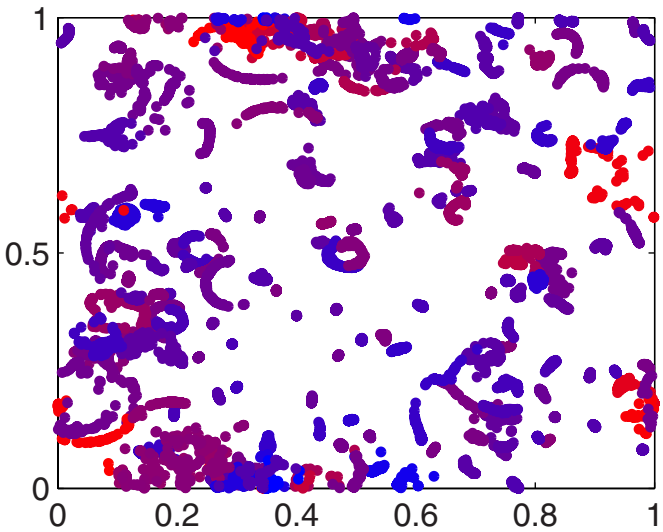

FIG. 3. (Color online) Stroboscopic plot of the particle position after each cycle for the 2D system of interacting particles. Plot after 100 cycles for a period $T=2$, same parameters as in Fig. 1 (open symbols). Only 150 trajectories are shown. Colors code for the particle diffusivity.

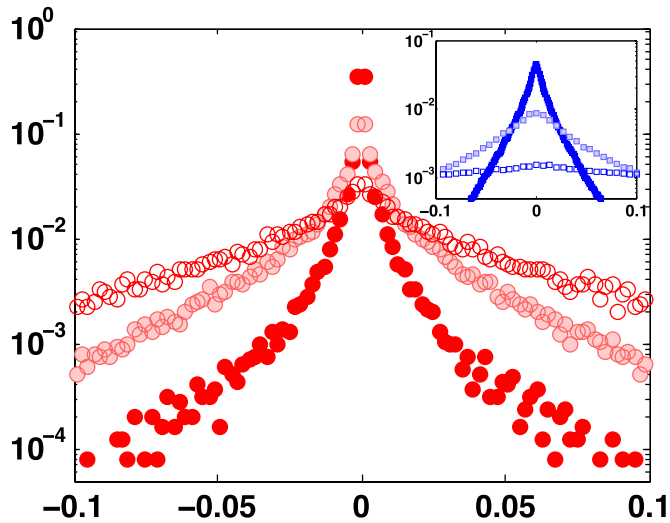

FIG. 4. (Color online) Probability density function of the particles diffusivity for $T=2$ (dark red circles), $T=2.2$ (light red circles), and $T=2.4$ (open circles). Same parameters as in Fig. 1 (open symbols). Inset: Same histograms for the coupled maps with $\epsilon=(1/\sqrt{2})\times 10^{-12}$ and $T=28$ (dark blue squares), $T=30$ (light blue squares), and $T=32$ (open squares).

mixing flow [20]. In such cases, the relative importance of the two effects cannot be disentangled by the measurement of the effective diffusivity of the particles and may be harder to assess experimentally.

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