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We revisit Taylor's experiment investigating the evolution of a blob of dye in a periodically sheared suspension of non-Brownian particles. Above a critical strain amplitude, particulate suspensions are subject to phase transition where reversibility is lost and particles fail to return to their original positions. We investigate the effect of this transition on the dispersion of a blob of dye. Beyond the critical strain, the dispersion of the blob is found to increase significantly. The dispersion coefficient of the blob of dye is measured and compared to the self-diffusivity coefficient of the particles.

In 1966, G.I. Taylor illustrated in a now famous experiment the reversibility of Stokes flow [1]. A blob of dye is initially injected in a Couette cell filled with a very viscous and Newtonian fluid. When rotating one of the cylinder, the fluid is linearly sheared and the blob of dye stretches into a thin filament until almost disappearing. Then, upon reversing the rotation for the same number of turns, the drop astoundingly recovers its initial shape (except for slight blurring due to molecular diffusion).

Inspired by Taylor's work, we performed the exact same experiment but in a suspension of non-Brownian particles to investigate how the presence of particles within the fluid breaks the reversibility of the flow and influences the dispersion process of the blob of dye. Under periodic shear, Pine *et al.* [2] demonstrated that non-Brownian suspensions undergo a dynamical phase transition. For a given particulate volume fraction and a small enough strain amplitude, the suspension relaxes to an absorbing reversible state in which particles return to their original position after every cycle of shear. However, when the strain amplitude exceeds a critical value γ_c, which depends on the volume fraction and the particle roughness [3], the suspension transitions to a fluctuating state: the particles do not return to their original positions and, when tracked stroboscopically at the end of each cycle of shear, the particles exhibit large fluctuations analogous to a random walk. In that fluctuating state, the loss of reversibility of the particles motion should also induce a loss of reversibility in the fluid motion. The goal in the present Letter is to perform Taylor's experiment in a particulate suspension to study the effect of this transition on the dispersion of the blob of dye.

Under continuous shear, the phenomenon called "shear induced-diffusion", which was widely investigated [3–10], was shown to significantly enhance the transfer of heat [9, 11] or mass [12, 13] across suspensions of non-Brownian particles. This enhancement occurs since under shear, particles within the fluid constantly collide with one another and change streamlines. The particles thus generate disturbances within the fluid which promote the dispersion of the scalar (temperature or concentration), prelude to its subsequent mixing. Particles also significantly affect transfers close to boundaries: the par-

ticle rotation was shown to disrupt the diffusive boundary layer by a 'rolling-coating effect' (Souzy *et al.* [13]) which convects the scalar at a constant rate from the wall, where it is injected, to the bulk of the suspension.

The dispersion enhancement of a scalar field has been already quite studied in suspensions undergoing a continuous shear flow, but it has yet never been studied under periodic shear. Our motivation is that this latter flow configuration is particularly appealing since, after each cycle of shear, the affine part of the flow (the reversible part) cancels out. Therefore, by measuring the evolution of the blob of ink stroboscopically, *i.e.* at the end of each cycle of shear, one can investigate the blob's dispersion solely focusing on the effect of the fluctuations induced by the particles, not having to account for the background linear stretching as in a continuous shear.

An original experimental setup was developed to perform direct visualizations of the dispersion of a blob of dye in a periodically sheared suspension of non-Brownian particles. By measuring the diffusion coefficients of both the particle and the fluid phases, we find that the blob's dispersion follows the same dynamical phase transition as the particles.

The experimental setup, shown in figure 1(a), consists of a transparent cell within which a transparent mylar belt, mounted tightly on two vertical rotating cylinders, generates a linear shear flow. A precision rotation stage drives the rotation of one of the vertical cylinders. The fluid is a Newtonian mixture of Triton X-100 (73.86 wt %), zinc chloride (14.24 wt %) and water (11.90 wt %), identical to that used in Souzy *et al.* [13]. Its viscosity $\eta = 3$ Pa s is large enough to ensure a viscous flow ($Re \approx 10^{-4}$) and its composition was chosen to match both the particle density ($\rho_f = \rho_p = 1.19$ g.cm^{-3}) to avoid sedimentation, and their optical index of refraction. A small amount of hydrochloric acid (≈ 0.05wt%) is added to the solution to prevent the formation of zinc hypochlorite precipitate, thereby significantly improving the optical transparency of the solution. The temperature of the setup is also adjusted ($\pm 0.05°$C) with a water-bath jacket connected to a cryo-thermostat in order to finely tune index-matching between the fluid and the particles. We use mono-disperse spherical PMMA beads from
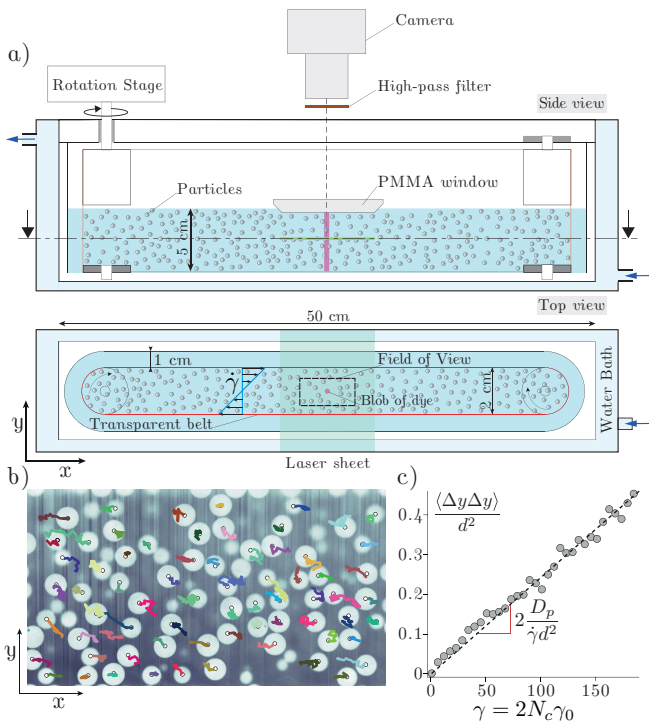
FIG. 1. a) Schematic of the setup, top and side views. b) Image of the suspension seen from the top (particles appear as white discs) and stroboscopic particle trajectories for cycles of shear of strain amplitude $\gamma_0 = 2.8$. c) Corresponding particle mean square displacement versus total accumulated strain $\gamma = 2N_c\gamma_0$.

Engineering Laboratories Inc. with a diameter $d = 2$ mm which were chosen for their high surface quality and good transparency. Note that, above the region of interest, a small plexiglass window is placed on the free surface of the suspension to avoid its deflection during shear which would deteriorate the visualization. A small hole, through the window, is used to inject the blob of dye in the middle of the suspension with a thin needle. The molecular diffusion coefficient of the dye in the fluid at rest is $D_0 \approx 10^{-13}\mathrm{m}^2\mathrm{s}^{-1}$ [14]. All the experiments were done at a volume fraction of $\phi = 35\%$.

Two types of experiments were performed i) to follow the evolution of the blob of dye, the suspension is perfectly transparent and the blob consists in a small injection of fluid doped with a fluorescent tracer (rhodamine 6G) , ii) to track the particle motion, the whole fluid is homogeneously doped with the fluorescent dye. In both cases, visualization is performed from the top of the cell by imaging the rhodamine which fluoresces under illumination provided by a thin horizontal laser sheet ($\approx 60\mu m$ thick). Images are acquired using a high resolution camera (Nikon D-300S). A high-pass filter ($\lambda > 590$ nm) is used to eliminate direct light reflexions and collect the light solely emitted by the fluorescent rhodamine.

As presented in figure 1(b), when the whole fluid is doped with rhodamine, the particles within the light sheet appear as white discs, while the fluid is dark. Note that all the particles have the same size; the apparent size differences arises from their different vertical positions relative to the light plane. Images are first thresholded, then an algorithm of particle detection based on the circular Hough transform [9, 15] is applied to find the center location of each particle within ± 1 pixel. Particles trajectories are then reconstructed by applying a tracking algorithm. A very important point is that for an experiment where periodic oscillations are performed at the strain amplitude γ_0, prior to injecting the blob or measuring the particle trajectories, the suspension is first pre-sheared at that strain amplitude for 40 cycles in order to let the suspension self-organize into its steady-state configuration [16]. To track the particles, images are taken continuously every strain increment of 0.1 while for blob experiments, images are only acquired at the end of each cycle of shear. A cycle of shear consists in slowly shearing (at 0.1 s^{-1}) the suspension forward for a strain γ_0, then shearing it backward to the initial position. For each experiment, N_c cycles of shear are performed and the total accumulated strain is thus $\gamma = 2N_c\gamma_0$.

We first present results characterizing the dynamics of the particles. Trajectories of the particles are recorded continuously to be able to track particles from one cycle to the next. However, to quantify the amount of irreversibility, we compute the mean square displacement of the particles only using their positions at the end of each cycle. An example of such trajectories is represented in figure 1(b) for an experiment performed at large strain amplitude. The mean square displacement in the gradient ($y-$) direction $\langle \Delta y \Delta y \rangle$, where $\Delta y = y(t) - y(t=0)$ and the angle brackets denote an average over all particles, is close to zero for small applied strain amplitudes. This means that the particle motion is reversible. However, for experiments performed at larger strains, the mean square displacement increases linearly with time indicating that the particles do not return to their original position and follow a diffusive behavior, see figure 1(c). Following Pine *et al.*, the slope of the particle mean square displacements is used to define a dimensionless particle diffusion coefficient $D_p^* = \langle \Delta y \Delta y \rangle/(2d^2\dot{\gamma}t)$ which quantify the irreversible motion of the particles. We recover the results of Pine *et al.* that particles, beyond a critical strain amplitude of $\gamma_c \approx 2$ for the present volume fraction $\phi = 35\%$ (equivalent of $\gamma_c \approx 1$ with the definition of the strain amplitude used by Pine [2]), transit to a fluctuating state, see figure 4.

We now investigate the effect of this transition on the dispersion of a blob of dye. After pouring the suspension (without rhodamine) within the cell and pre-shearing it, a cylindrical blob of dyed fluid is injected at $t = 0$ and with the fluid at rest in the middle of the shear cell. The blob has an initial diameter $s_0 \simeq d$ and is aligned with the vorticity direction of the shear flow and centered on
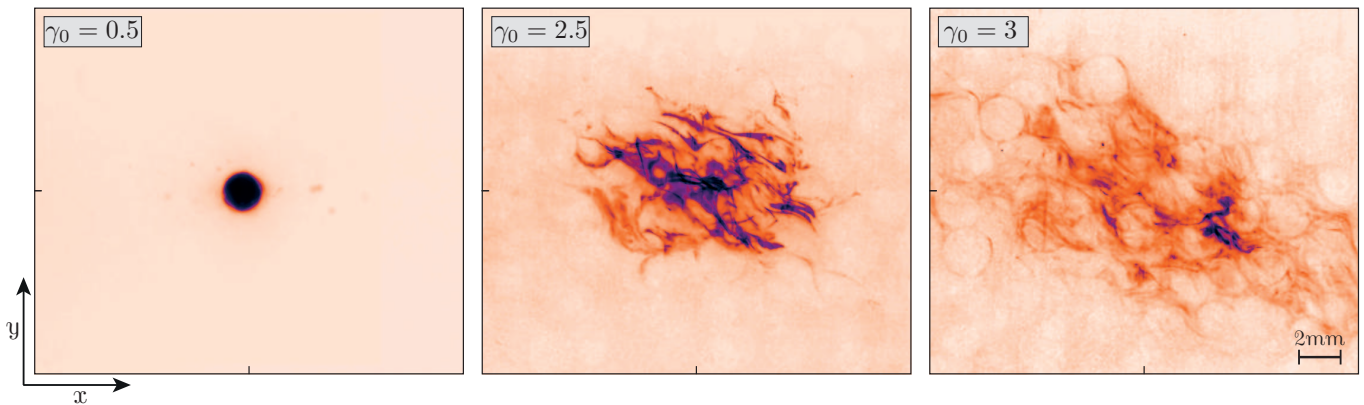
FIG. 2. Image of the blob after 40 cycles of shear performed at a strain amplitude : $\gamma_0 = 0.5$, $\gamma_0 = 2.5$, $\gamma_0 = 3$. Each image corresponds to the superposition of 10 pictures taken from 10 independent experiments performed at the same strain amplitude, see also Movie 1 and Movie 2.
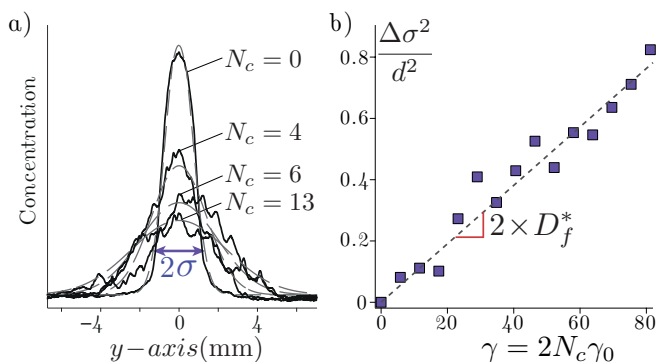

FIG. 3. a) Successive concentration profiles for a blob submitted to a strain amplitude $\gamma_0 = 2.9$. The profiles are obtained by averaging over 10 independent runs. The dashed curves correspond to the best gaussian fit. b) Evolution of the corresponding dimensionless relative variance $\Delta\sigma^2/d^2$ as a function of the total accumulated strain $\gamma = 2N_c\gamma_0$.

the neutral velocity plane. This results in a macroscopically two-dimensional initial configuration, and ensures that the blob does not drift with the flow. The concentration of rhodamine within the blob was adjusted so that absorption of the light intensity can be neglected. The suspension is then slowly sheared back and forth over $N_c = 40$ cycles at the strain amplitude γ_0.

Figure 2(a, b, c) shows how the blob has evolved after 40 cycles of shear for experiments performed at different strain amplitudes. Note that each image is the superposition of pictures taken from 10 independent experiments performed with the same imposed strain amplitude. At small strain amplitude $\gamma_0 = 0.5$, as in Taylor's experiment, the blob after each cycle of shear recovers its initial shape, see figure 2(a). Conversely, for larger applied strain amplitudes, the blob rapidly spreads in the surrounding medium, evolving towards a topology constituted of filament-like structures [17] (to visualize the continuous evolution of the blob, see Movie 1 and Movie 2). Larger strain amplitudes result, after the same num-

ber of cycles, into a larger dispersion of the blob.

More quantitatively, we can characterize the blob effective dispersion by examining its spreading. Figure 3(a) shows successive concentration profiles obtained by averaging the light-intensity level along the x-direction. The low concentration of rhodamine used for the experiments ensures a linear relation between light intensity and dye concentration. Two important observations can be made. First, the concentration distribution is normally distributed. The dashed curves are the best gaussian fit where, once the amplitude of the initial distribution (for $N_c = 0$) is fixed, the sole remaining adjustable parameter is the standard-deviation σ. Second, the relative variance of the concentration distribution $\Delta\sigma^2(t) = \sigma^2(t) - \sigma^2(t=0)$ increases linearly with the total accumulated strain $\gamma = 2N_c\gamma_0$ (equivalent to time), see figure 3(b). These two features evidence that the dispersion process of the blob, and thus of the interstitial fluid phase in between the particles, is diffusive. The effective dispersion coefficient of the fluid phase is provided by the classical definition

$$\Delta\sigma^2 = 2D_f t. \tag{1}$$

The dimensionless effective dispersion coefficient of the fluid phase, D_f^*, can be defined by the dimensionless form of equation 1 ($\Delta\sigma^2/d^2 = 2\frac{D_f}{\dot\gamma d^2}\gamma$) as $D_f^* = D_f/\dot\gamma d^2$ which corresponds to one half the slope of the linear fits presented on figure 3(b). Error bars are also estimated from the typical standard deviation between runs, see figure 4.

Similarly to the behavior of the particles, for the volume fraction $\phi = 35\%$ investigated, above a critical strain $\gamma_c \approx 2$, the dispersion coefficient of the blob goes through an abrupt transition, see figure 4 where D_p^* and D_f^* are plotted versus the applied strain amplitude γ_0. The loss of reversibility induced by the presence of the particles significantly accelerates the dispersion process. Indeed, if
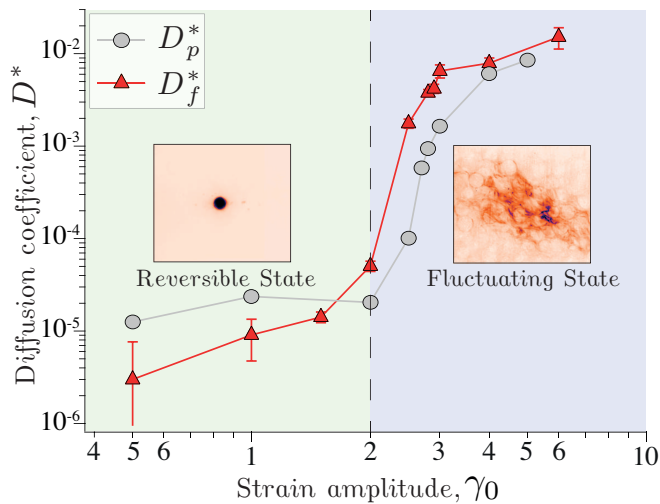
FIG. 4. Dimensionless self-diffusivity coefficient of the particle D_p^* and dimensionless dispersion coefficient of the fluid D_f^*, as a function of the applied strain amplitude γ_0. Reversibility is lost above a critical strain amplitude $\gamma_c \approx 2$.

one was only considering the effect of molecular diffusion, *i.e.* if the blob was sheared in a pure fluid without particle, the transverse dispersion coefficient of the dye would be $D_f = D_0$, leading to a dimensionless effective dispersion coefficient of the fluid phase, $D_f^* = D_0/\dot{\gamma}d^2 = 10^{-6}$. In the fluctuating state, (beyond γ_c), the dimensionless dispersion coefficient of the fluid is $\approx 10^{-2}$ so about 4 orders of magnitude larger than that of molecular diffusion alone. Note that, as one can guess from figure 2, the dispersion coefficients in the flow direction (not shown here) follow very similar trends as those in the gradient direction.

For $\gamma_0 < \gamma_c$, in the so-called reversible state, we measure effective diffusion coefficients of $\approx 10^{-5}$ both for the particles and for the blob. For the particles, which are non-brownian, D_p^* should vanish when $\gamma_0 \to 0$. This small discrepancy arises from experimental artefacts which are noise from the Hough detection algorithm (detection of the particle center locations within ± 1 pixel) and unavoidable slight sedimentation or natural convection currents within the fluid. In that limit, the latter two effects probably also bias the measurement of the blob dispersion coefficient. It is noteworthy that the measured self-diffusivity coefficient of the particles obtained at large strain amplitudes are in good agreement with previous measurements done in continuously sheared particulate suspensions [7, 9]. Similarly, the enhancement of the dye diffusion coefficient at large applied strain amplitude is of the same order $\approx 10^4$ than in a continuously sheared suspension [13]. This result could be useful in certain mixing applications where oscillatory shear might be easier to implement than continuous shear [18].

The molecular diffusion coefficient of rhodamine 6G in the present suspending liquid is small ($D_0 \approx 10^{-13}\text{m}^2.\text{s}^{-1}$). In such condition of large Péclet number, $Pe = \dot{\gamma}d^2/D_0 = 10^6$, (with $s_0 \simeq d$) where the effect of molecular diffusion is negligible relative to the fluctuations induced by the presence of the particles [5, 19], the measurement of the dye effective dispersion coefficient provides a good estimate of the fluid dispersion coefficient. Conversely to what was reported by Breedveld *et al.* [20], we find that the fluid dispersion coefficient is always slightly larger than that of the particles, see figure 4. One can explain this trend by the fact that the self-diffusivity coefficient of the particles only accounts for their translational diffusivity, while the fluid dispersion coefficient arises from the perturbations induced by both the translational and the rotational motion of the particles.

We performed Taylor's experiment in a suspension of non-Brownian particles to investigate how the loss of reversibility induced by the presence of the particles affects the dispersion of the blob of dye. By measuring both the particles and the blob of dye transverse effective diffusion coefficients, we found that the dispersion of the blob undergoes the same transition as the one observed by the particles [2]. At small strain amplitude, as in Taylor's experiment, the system is reversible and the blob thus recovers its initial shape after each cycle of shear. For a volume fraction $\phi = 35\%$, beyond a critical strain amplitude $\gamma_c \approx 2$, the system transits to a fluctuating state where reversibility is lost. As a result, the blob spreads very rapidly. The blob dispersion is found to be diffusive with an effective diffusive coefficient of the same order as the self-diffusivity coefficient of the particles.

[1] G.I. TAYLOR, & J. FRIEDMAN "Low Reynolds Number Flows," *Nat. Com. on Fluid Mech. Films*, (Encyc. Brit. Educ. Corp.), United States (1966).

[2] D. J. PINE, J. P. GOLLUB, J. F. BRADY & A. M. LESHANSKY "Chaos and threshold for irreversibility in sheared suspensions," *Nature*, **438(7070)**, 997–1000 (2005).

[3] P. PHAM, J.E. BUTLER & B. METZGER "Origin of critical strain amplitude in periodically sheared suspensions," *Phys. Rev. Fluids*, in Press. (2016).

[4] P. A. ARP, AND S. G. MASON, "The Kinetics of Flowing Dispersions IX. Doublets of Rigid Spheres (Experimental)," *Journal of Colloid and Interface Science* **61**, Issue 4095, 44–61 (1976).

[5] E. C. ECKSTEIN, D. G. BAILEY, AND A. H. SHAPIRO, "Self-diffusion of particles in shear flow of a suspension," *J. Fluid Mech.* **79**, 191–208 (1977).

[6] F. R. DA CUNHA, AND E. J. HINCH, "Shear-induced dispersion in a dilute suspension of rough spheres," *J.*

Fluid Mech. **309**, 211–223 (1996).

[7] Sierou A. & Brady J. F. 2004 Shear-induced self-diffusion in non-colloidal suspensions, *J. Fluid Mech.* **506**, 285314.

[8] F. Blanc, F. Peters, & E. Lemaire "Experimental signature of the pair trajectories of rough spheres in the shear-induced microstructure in noncolloidal suspensions," *Phys. Rev. Lett.*, **107(20)**, 208302 (2011).

[9] B. Metzger, O. Rahli, & X. Yin "Heat transfer across sheared suspensions: role of the shear-induced diffusion," *J. Fluid Mech.*, **724**, 527–552 (2013).

[10] P. Pham, B. Metzger, & J.E. Butler "Particle dispersion in sheared suspensions: Crucial role of solid-solid contacts," *Phys. Fluids*, **27(5)**, 051701 (2015).

[11] Sohn C. W. & Chen M. M. Heat Transfer enhancement in laminar slurry pipe flows with power law thermal conductivities, *J. Heat Trans.* **106**, 539542. (1984).

[12] Wang N. L. & Keller K. H. Augmented Transport of Extracellular Solutes in Concentrated Erythrocyte Suspensions in Couette Flow, *J. Col. Int. Sc.* **103**, 210225. (1985).

[13] M. Souzy, X. Yin, E. Villermaux, C. Abid, & B. Metzger "Super-diffusion in sheared suspensions," *Phys. Fluids*, **27(4)**, 041705 (2015).

[14] C. T. Culbertson, S. C. Jacobson, & J. M. Ramsey "Diffusion coefficient measurements in microfluidic devices," *Talanta*, **56**, 365–373 (2002).

[15] T. Peng "Detect circles with various radii in grayscale image via Hough Transform," http://www.mathworks.com/matlabcentral/fileexchange/9168-detect-circleswith-various-radii-in-greyscale-image-via-hough-transform.

[16] L. Corté, P. Chaikin, J. P. Gollub, & D. J. Pine "Random organization in periodically driven systems," *Nature Physics*, **4(5)**, 420–424 (2008).

[17] J.M. Ottino "The Kinematics of Mixing: Stretching, Chaos, and Transport," *Cambridge University Press* (1989).

[18] M. Workamp, V. Saggiomo & J. A Dijksman "A simple low pressure drop suspension-based microfluidic mixer," *J. Micromech. Microeng.*, **25**, 094003 (2015).

[19] D. Leighton, and A. Acrivos, "Measurement of shear-induced self-diffusion in concentrated suspensions of spheres," *Journal of Fluid Mechanics* **177**, 109–131 (1987).

[20] V. Breedveld, D. van den Ende, A. Tripathi, & A. Acrivos "The measurement of the shear-induced particle and fluid tracer diffusivities in concentrated suspensions by a novel method," *J. Fluid Mech.*, **325**, 297–318 (1998).