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Reversible self-organisation of a sheared suspension

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Flows at low Reynolds numbers are driven by time reversible equations. However, experiments showed that sheared suspensions of micro-scale particles in a viscous fluid do not always exhibit a reversible behaviour: the particles do not come back to their initial position after a back-and-forth movement. This study provides some possible answers to this paradox comparing experimental data to computer model results. This behaviour can be explained by irreversible short range interactions between particles and depends largely on the shearing amplitude. For low amplitudes, the system first self-organises in a stable configuration. Such a configuration minimises particles interactions during the sheared movement. Therefore the system becomes totally reversible. When the shearing amplitude increases the relative movement between particles increases as well and so does the probability of interaction. Above a critical amplitude, the system cannot reach any stable configuration. It becomes chaotic and does not self-organise. It exhibits a phase transition dynamics.

1. Introduction

About a century ago, G.I Taylor [1] showed that the motion of a fluid can be reversible. If a viscous fluid is placed between two concentric cylinders, the rotation of the inner cylinder will shear the liquid. However, if it is rotated in the other direction, the motion of the liquid will be exactly inversed. Thus, if a coloured drop is placed in this liquid, the first rotation will spread the drop. Yet when the cylinder is rotated in the other direction, the coloured drop will reform (see Figure 1).

This spectacular and non-intuitive observation can be explained by laws of motion. Hydrodynamics motion is driven by the non-linear and irreversible Navier-Stokes equation. Consequently a dash of milk will never reform. However, if the liquid is viscous enough, this equation is simplified and becomes reversible. Thus, for an incompressible fluid at low Reynolds numbers ($\mathrm{Re} = \rho\frac{\mathrm{UL}}{\eta} << 1$) Stokes equations become:

$$\boldsymbol{\nabla}.\boldsymbol{v} = 0$$

$$-\boldsymbol{\nabla}P + \eta\triangle\boldsymbol{v} = 0$$

where P is the pressure, η the shear viscosity and $\boldsymbol{v}$ the liquid velocity. At the boundary, the liquid velocity is equal to the wall velocity. These equations give reversible solutions and a reversal of the boundary motion would immediately invert the flow velocity in every points. Hence a viscous liquid sheared between two concentric cylinder is a reversible system.

Based on this result, a suspension of macroscopic particles in such a fluid should a priori be reversible. After a back-and-forth rotation of the inner cylinder, all the particles should return to their initial position. However, in 2010, D. G. Pine et al [2] showed that a slowly sheared suspension of solid particles at low Reynolds number have an irreversible behaviour if the shearing amplitude exceeds a critical value. There is a symmetry breaking that occurs through a critical phenomenon. This cannot be explained by the study of only one particle but is caused by particles interactions.

This study deals with the amplitude of these micro-

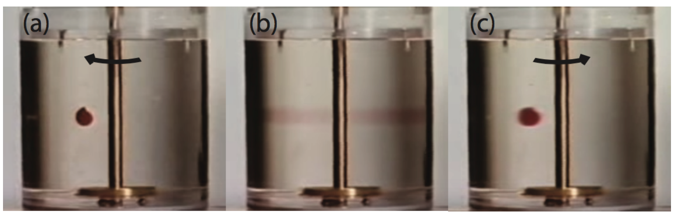

Fig. 1 Pictures of the Taylor experiment on the reversibility at low Reynolds numbers. (a) Pigments are placed in a viscous fluid between two concentric cylinders. (b) The inner cylinder is slowly rotated 4 turns forwards. (c) The inner cylinder rotated back 4 turns. The pigments return to their initial position. The flow is reversible. Pictures from "Low-Reynolds-Number Flows" lecture by National Science Foundation [1].

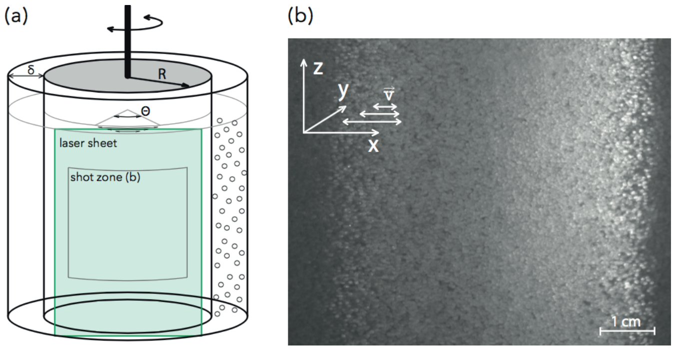

Fig. 2 (a) Setup for a viscous suspension under an oscillatory shear in a Couette geometry for a shearing amplitude γ. (b) Picture of suspended particles in a laser sheet lighting the gap between the two cylinders.

scopic interactions and how a statistical description of these interactions can explain such a critical transition. The first part of this paper exposes the phase transition behaviour of a micro-scale particles suspension in a sheared Couette flow at low Reynolds number. The second part of this article explains a simple numerical model of the phenomenon. This model suggests an amplitude for particles interactions and a physical interpretation of this phase transition behaviour.

2. Experiments

Design. In order to study the behaviour of sheared suspension, UCON™ oil and micro-scale polystyrene particles are mixed together. The suspension is placed between two concentric cylinders separated by a gap $\delta = 5$ mm. The inner cylinder has a radius R = 50 mm. The particles diameter is 100 μm and the volume ratio ($V_{\text{particles}}/V_{\text{total}}$) is fixed at 0.2. In order to shear the fluid, the inner cylinder is subsequently rotated by an angle $\Theta \ll 2\pi$. This movement corresponds to a shearing amplitude $\gamma = R\Theta/\delta$. The rotation speed is chosen in accordance with the UCON™ oil viscosity (about 15 Pa.s at 293 K) to keep the Reynolds number very low (Re $< 10^{-4}$). Because the density ratio between polystyrene and UCON™ oil is 0.93 at 293 K, sedimentation is extremely slow: about 0.05 μm.s^{-1}. During the experiments, the minimal displacement observed during 1 s is 0.3 μm. Therefore any movements induced by sedimentation will be neglected. Before each measurement, the inner cylinder is quickly rotated in one direction during about 50 turns in order to randomise the particles distribution. During the measurement, it is slowly and precisely moved using a stepper motor (ORIENTAL MOTOR U.S.A. Corp., model AR98MAD-H100-3). This motor as an angular resolution of 0.0036 ° by step yielding a precision on the shearing amplitude: $\Delta\gamma = 6.10^{-4}$. Each time the inner cylinder comes back to its initial position, the particles displacement is analysed to evaluate the reversibility. For that purpose, the suspension is lit up with a vertical laser sheet in order to observe the particles displacement. The area is filmed with a CCD camera (Thorlabs, model DCU224C, 1280 x 1024 Pixels),(See Figure 2). On these pictures a pixel corresponds to about 5 μm. The camera is programmed to take a picture each time the inner cylinder comes back to its initial position. The cylinder makes between 40 and 100 back-and-forth movements – called a cycle – for each shearing amplitude. Thus, a stroboscopic film of the system is obtained. This measurement is done for five different shearing amplitudes: $\gamma = 0.25$, 0.50, 0.75, 1.50 and 2.50.

In order to characterise the average particles displacement, two overlaps are calculated.
The first one, $O_\gamma(n)$, compares each picture to the previous one. It represents the decorrelation between two consecutive images. This characterises the average amplitude of the particles displacement after one cycle. It is defined by:

$$O_\gamma(n) = \frac{1}{Z_\gamma}\sqrt{\sum_{\text{pixels}(x_i,y_i)}\left[\frac{I_n(x_i,y_i)}{M_n} - \frac{I_{n-1}(x_i,y_i)}{M_{n-1}}\right]^2}$$

$I_n(x_i, y_i)$ is the intensity of the (x_i, y_i) pixel of the picture n. M_n is the average intensity of the n picture correcting global intensity fluctuations. Z_γ is a normalisation coefficient. It corresponds to the maximal decorrelation that could be observed on this measurement. It is calculated taking different images of the system in random configurations and corresponds to the average decorrelation between these pictures.
The second overlap, $\bar{O}_\gamma(n)$, compares each picture to a reference picture. It represents the decorrelation between an image n (for $n \geq 10$) and image 10 – when the system is in a permanent regime – and characterises the average amplitude of the particles accumulated displacement. It is defined by:

$$\bar{O}_\gamma(n) = \frac{1}{Z_\gamma}\sqrt{\sum_{\text{pixels}(x_i,y_i)}\left[\frac{I_n(x_i,y_i)}{M_n} - \frac{I_{10}(x_i,y_i)}{M_{10}}\right]^2}$$

The smaller these quantities the more similar the pictures. That is to say, the particles return to their previous positions. On the contrary, when particles do not return to their previous positions, these quantities tend to 1.

Results & Phase Transition. For the two highest amplitudes ($\gamma = 1.50$ and 2.50), the behaviour of the system is the same (See Figure 3 (a)). $O_\gamma(n)$ fluctuates widely: between 0.3 and 0.6 for $\gamma = 1.50$ and between 0.5 and 0.7 for $\gamma = 2.50$. See 3 (a) and (c). After each cycle the configuration of the particles changes: every particles position changes. $\bar{O}_\gamma(n)$ increases and saturates around 0.9 within 20 cycles (See Figure 3 (b)). So after 20 cycles, the system is in a complete different configuration.

For these amplitudes, the average particles displacement is estimated by PIV around 4 μm per cycle (See 4 (b)). This displacement is not organised. A PIV done between the cycle 30 and 40 for the shearing amplitude $\gamma = 1.50$ shows that the particles go in every direction (See 4 (a)). This measurement is done on 10 cycles in

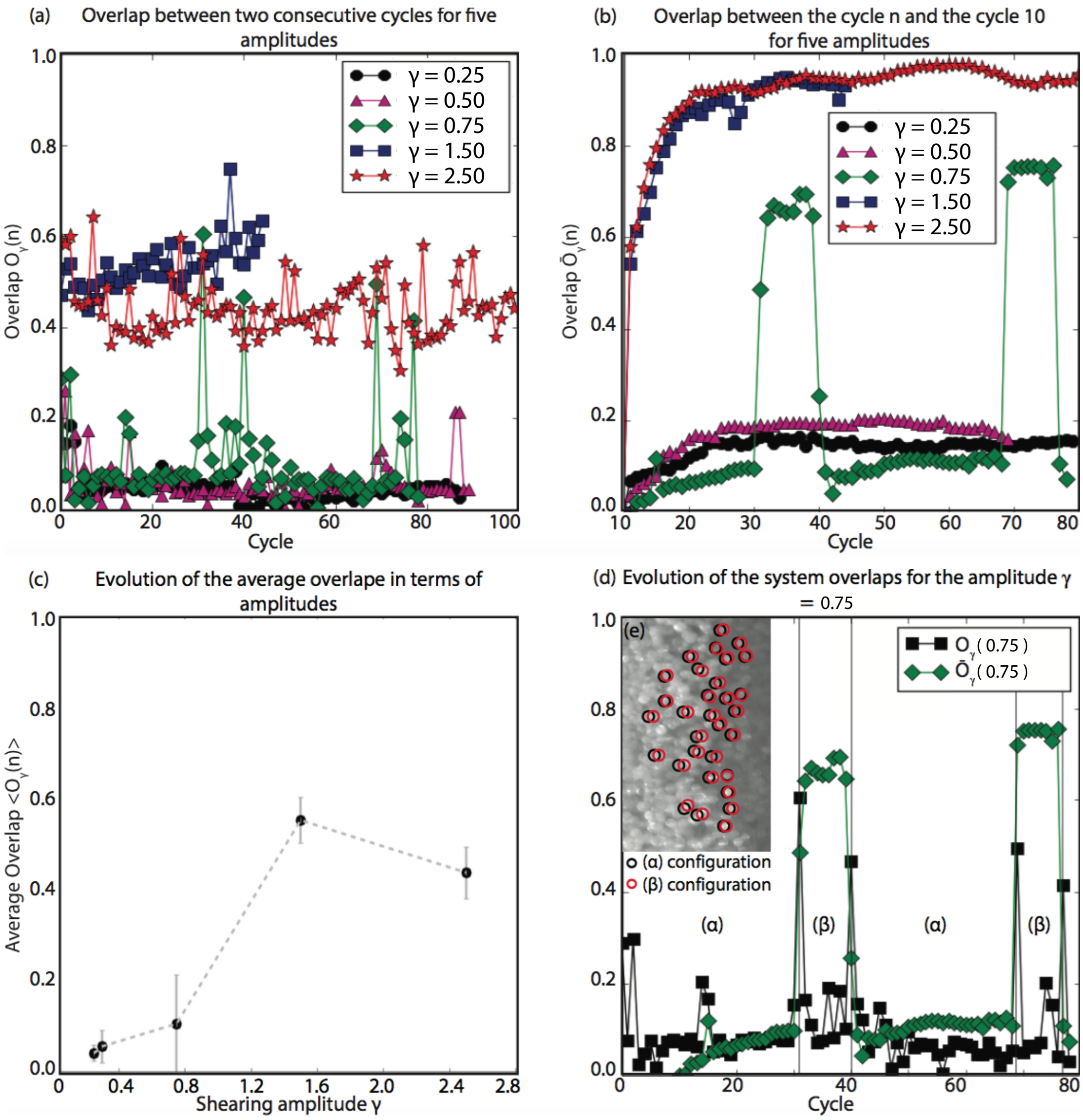

Fig. 3 (a) Overlaps between the picture of the system taken at the cycle n and a picture taken at the cycle $(n-1)$. (b) Overlaps between the picture taken at the cycle n and the picture taken at the cycle 10. (c) Average overlap between consecutive pictures for different shearing amplitudes. (d) Overlaps for the amplitude $\gamma = 0.75$ between a picture taken cycle n and the picture taken at the cycle 10 and between a picture taken at the cycle n and a picture taken at the cycle $n-1$. The system switches between two configurations (α) and (β). (e) Position of some particles in the configuration (α) in black and in the configuration (β) in red.

order to accumulate enough displacement.

For the two lowest amplitudes ($\gamma = 0.25$ and 0.50), $O_\gamma(n)$ is much lower and more stable. It fluctuates between 0 and 0.3 during the first 20 cycles and then between 0 and 0.09 (See figure 3 (a) and (c)).However $O_\gamma(n)$ is not zero: a slight movement is observed but its average amplitude estimated by PIV is only about 0.4 μm per cycle (See 4).
After 30 cycles, $\bar{O}_\gamma(n)$ remains low as well (below 0.2) (See Figure 3 (b)). It is not zero because of the slight movement observed previously which is accumulated over 30 cycles.
Yet, it does not increase more than 0.2 because this movement amplitude is too low to mix the particles positions. The particles only move around their initial positions. The global configuration does not change: it is stable. However, during the first cycles, $O_\gamma(n)$ is much higher. For $\gamma = 0.25$, $O_{0.25}(n)$ starts at 0.15 and decreases below 0.05 after the cycle 4. During the first 4 cycles the system configuration changes and organises itself in order to reach a stable configuration, that is to say a configuration in which the system is reversible. For $\gamma = 0.50$, the transition is less obvious. However, during the first 15 cycles, $O_{0.50}(n)$ fluctuates more than after the cycle 16 with amplitude variations decreasing from 0.3 to 0.1. The system takes more time to reach a stable configuration.

These measurements are consistent with the Pine *et al.* observations [2]. A cyclically sheared suspension at low Reynolds numbers presents a phase transition. The nature of the system depends on the shear stress amplitude and presents a critical amplitude. More measurements have to be done in order to completely characterize this phase transition. Below this critical amplitude the system reaches a stable and reversible configuration. The average movement of particles is too low to change the system configuration. After the critical amplitude, the average movement is too high for the system to find any stable configuration : the system is chaotic.

Phase Transition Nature. The shearing amplitude $\gamma = 0.75$ presents a threshold. During the first 15 cycles, the system organises itself the same way as it does for $\gamma = 0.50$ (See figure 3 (a) and (d)). However $O_\gamma(n)$ peaks between cycles 30 and 31, 40 and 41, 69 and 70, 77 and 78: each time the system appears to be in a different configuration. This observation is correlated with $\bar{O}_{0.75}(n)$ that increases from 0.05 to 0.18 (See figure 3 (c) and (d)). The two different values of $\bar{O}_{0.75}(n)$ correspond to two different configurations α and β (See Figure 3 (d)). Although perfect synchronisation with the camera was ensured, this is probably a measuring error. For instance the motor could miss a step in the first part of a cycle. This would shift the whole rotation during several cycles. Then, if the motor misses another step in the second part of a cycle, this would shift the whole rotation back.

However, an interesting physical explanation could be proposed. After the first 10 cycles, the system could reach a stable configuration α. Then between cycles 30 and 31, the system would change its configuration to reach another stable configuration β and would remain in this configuration during 10 cycles. After this switch, the decorrelation between two following cycles would drop back but the decorrelation with the cycle 10 (See Figure 3 (d)) in the configuration α would remain high. Between cycles 30 and 31, the system would change its configuration again and return in the configuration α. Thus the decorrelation with the image 10 would decrease. The fact that the system remains for longer in the α configuration than in the β configuration could indicate that the α configuration is more stable than the β configuration.

Thus, the movement amplitude could be high enough to create short-range-interaction instabilities. These instabilities would make the system change its configuration. The life time of these configurations being different, they could have different degrees of stability. Such an observation implies that the system follows at least a second order phase transition which has never been observed. However, this observation was done on one single experiment. It is not enough to conclude. It would be very interesting to study the existence of these metastable configurations. The threshold could be statistically interpreted as a brutal evolution in both the number and the stability of reachable

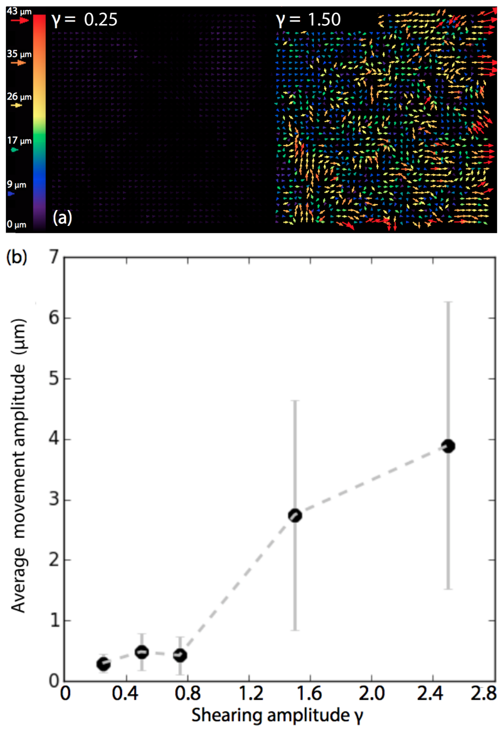

Fig. 4 (a) PIV between the pictures of the cycles 30 and 40 for the shearing amplitude $\gamma = 0.25$ and $\gamma = 1.50$. (b) Average movement amplitude between two consecutive cycles determined by PIV from the cycle 17 to 25 for the five different shearing amplitude.

distributions.

3. Numerical Model

Model. Based on a model developed by D.J. Pine *et al.* [2], this experimental system is simulated as a sheared flow between two planar boundaries. One of the plans moves in the X direction creating a simple shear flow with uniform velocity gradiant. The particles have a diameter of 100 μm and the volume ratio is set to 0.2 just as the experimental setup. The dimension of the simulated box is 5 mm by side and is adapted in order to simulate 500 particles. The particles movement is assumed to be equal to the movement of the fluid because of the low Reynolds numbers. The speed gradient is in the Y direction and the Z direction represents the height (See Figure 5 (a)). The boundary conditions in the X direction are periodical while the box's walls are impenetrable in the other directions.

At every step of the simulation, each particle moves between 0 and 100 μm according to speed gradient evaluated at its position. The value of the maximum step movement − 100 μm − has to remain small compared to the distance between particles. If two particles are in contact, their positions are randomly redistributed in a 100 μm diameter sphere around their position. Indeed interactions cannot be coded by elastic chocks or any other reversible interactions. If they were, all the process would always be reversible regardless of the amplitude. The model needs to introduce irreversibility. Thus randomly redistributing the particles in a 100 μm diameter sphere is a simple way to code 100 μm range irreversible interactions. This procedure is iterated until the maximum shearing amplitude is reached. This amplitude corresponds to the experimental shearing amplitude γ. Then the movement is reversed in order to complete the cycle. This cycle is repeated 100 times for different cumulated amplitudes $\gamma = 0.25$, 0.50, 0.63, 0.70, 0.82, 0.88 and 1.50.

Results. Figure 5 (c) presents the number of interacting particles for two different amplitudes $\gamma = 0.25$ and 1.50. This number is calculated at each step of the algorithm and corresponds to the number of moving particles. Hence it is naturally linked to overlap notion presented in the experimental part. At the beginning each simulation starts with a random distribution where about 200 particles are interacting. For the lowest amplitude $\gamma = 0.25$ the system self-organises within 10 cycles. The number of interacting particles drops to zero during an entire cycle (See Figure 5 (c)). From this point the system is completely reversible. It is in a stable configuration and no more particles are interacting despite the sheared movement. For the highest amplitudes $\gamma = 1.50$, the system does not self-organise and it cannot find any stable configuration. There are always interactions between particles.

The Figure 5 (d) presents the average number of interacting particles after 20 cycles for different shearing amplitudes from 0.25 to 1.50. For amplitudes lower than $\gamma = 0.70$ this number is zero. Within 20 cycles, the systems self-organise themselves and become totally reversible. On the contrary, for higher amplitudes, the number of interacting particles never reaches zero and increases with the amplitude. The system is irreversible.

These results match the experimental data. There is a critical amplitude that occurs around the same amplitude : $\gamma = 0.70$. Thus it seems that the amplitude of interaction is relevant. The complex interactions between particles that lead to irreversibility have an amplitude smaller than

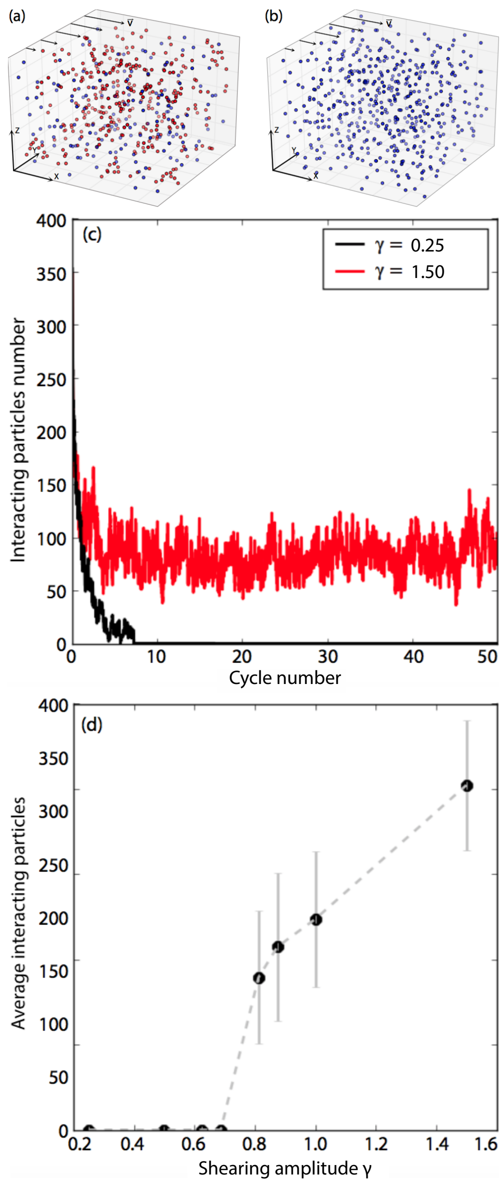

Fig. 5 (a) Initial random distribution of particles, interacting particles in red, non-interacting particles in blue. (b) Organised and reversible distribution of particles. (c) Number of interacting particles at every algorithm step over 50 cycles for shearing amplitudes of 0.25 and 1.50. (d) Average number of interacting particles after 20 cycles for different shearing amplitudes from 0.25 to 1.50. For shearing amplitudes lower than 0.70, the number of interacting particles goes to zero and the system reaches a totally reversible configuration. For higher amplitude the system never reaches a reversible configuration.

100 μm. The experimental behaviour around the critical amplitude – switching between two configurations – cannot be obtained with the model. Indeed, in the model, as soon as the system is in a reversible configuration, no perturbations can make it change.

This model mainly reproduces the experiment behaviour and hints towards an interpretation. Thus, in the experimental setup, the particles are probably interacting through the liquid with a amplitude smaller than 100 μm. However, this model uses random displacements as interactions. This does not correspond to real interactions. The question of the relevance of this model remains open.

4. Conclusion

Hydrodynamics equations imply that a system at low Reynolds numbers should always be time reversible. However, as observed by D. J. Pine et al [2], this experiment shows that a cyclic sheared suspension at low Reynolds numbers is not always reversible. The system presents a phase transition behaviour with a threshold in amplitudes. The value of the critical amplitude depends on characteristics such as density of particles.

When the system is sheared, the particles move because of the global reversible flow. Yet, if particles are close enough, they also interact together by short range interactions – contact or induced local flow – and move relatively to one another. The system is no longueur deterministic but has a stochastic behaviour. The numerical model indicates that these interactions have an amplitude lower than the particles diameter. At the beginning the particles distribution is random. Because of the shearing some particles become close enough to interact which leads to changes of configuration after each cycle. For shearing amplitudes lower than a critical amplitude γ_0, these changes bring the system in a stable configuration. In such a configuration the particles are organised in a way that minimises the interactions during a cycle. They only move because of the sheared flow. Thus, the system is reversible. For shearing amplitudes higher than γ_0, the particles movement during a cycle is significant enough to prevent any stable configuration. The system does not reach any organisation that avoids short range interactions during a whole cycle. Indeed, increasing the shearing amplitude increases the particles relative movement as well as the probability of short range interactions. Thus the particles interact and the system is not reversible. This dynamics implies that the critical amplitude increases with the average distance between particles, when the particles density decreases. Indeed increasing the average distance between particles reduces the probability of short-range interactions and creates stable configurations for higher amplitudes. This observation was done by D. J. Pine et al [2].

Our study could have revealed a new phenomenon. For an amplitude just below the critical amplitude γ_0, the system seems to switch between different stable configurations. However, more observations of this kind have to be done in order to conclude. It would be very interesting to study the existence of these metastable configurations.

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