PHYSICAL REVIEW E **81**, 062501 (2010)

Rheology and ordering transitions of non-Brownian suspensions in a confined shear flow: Effects of external torques

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(Received 12 April 2010; published 21 June 2010)

We investigate the effect of an external torque, applied in the vorticity direction, to particles in a sheared non-Brownian suspension confined by rigid walls. At volume fractions of $\phi=0.48-0.52$ such suspension flows undergo an ordering transition, developing a hexagonal structure of particle strings in the velocity gradient-vorticity plane. The hexagonal structure is disturbed by negative torques, leading to an increase in the shear viscosity. Positive torque has a favorable effect on the ordered state. However, if the magnitude of the positive torque exceeds a certain threshold, the hexagonal order begins to be weakened. Due to the significant changes in suspension microstructures, rheological parameters such as the shear and vortex viscosities exhibit nonlinear responses to the external torques. On the other hand, at lower volume fractions $\phi\leq 0.40$, where ordered structures are not developed, suspension microstructure is not sensitive to an external torque and the apparent viscosity is a linear function of the torque.

DOI: 10.1103/PhysRevE.81.062501 PACS number(s): 83.10.Tv, 47.57.E−, 47.57.Qk

Manipulating rheological properties of suspension flows by imposing electric or magnetic fields is of current interest due to its relevance to a wide range of technological applications [1]. In electrorheological (ER) or magnetorheological (MR) fluids, a dramatic increase in the apparent viscosity is observed when particles are assembled into well-organized large-scale structures, such as chains or stripes, by applied fields [2]. It is also known that the apparent viscosity can be reduced by applying an external torque on the particles in ferrofluids [3]. In ER fluids, such a decrease in apparent viscosity can be achieved by a dc electrorotation of suspended particles (Quincke rotation) [4]. If a dc electric field is applied in the velocity-gradient direction on suspensions under a steady shear, the particles experience an electric torque in the vorticity direction, which makes the particles act as a colloidal motor [5,6]. Lemaire *et al.* [6] suggested a simple model to predict the apparent viscosity of sheared suspensions under Quincke rotation. However, the detailed microstructure and rheological behavior of sheared suspensions under an external torque are not fully understood.

Rheological properties of suspension flows, such as the apparent viscosity and the normal stress differences, are closely related to the microstructure of the suspension. For example, in suspensions of non-Brownian particles under a steady shear, irreversible effects introduced by small roughness elements on the particle surface, residual Brownian force, and/or surface charge result in an anisotropic microstructure, which is responsible for the non-Newtonian rheology [7]. In recent studies of the colloidal [8] and non-Brownian suspensions [9,10] under a steady shear, an ordering transition is observed at high volume fractions ($\phi \simeq 0.50$). This nonequilibrium phase transition of non-Brownian particles is different from the thermodynamic phase transition of colloidal particles in that the process is driven mostly by the shear-induced hydrodynamic interactions between particles. As the suspension undergoes the ordering transition, the apparent viscosity decreases significantly. Particularly, Yeo and Maxey [10] showed that the ordering transition, and hence, the changes in the apparent viscosity are complex functions of the ratio of the channel height to the particle radius and the volume fraction.

In this Brief Report, we show a possibility of manipulating the ordering transition of non-Brownian suspensions in a Couette flow by applying external torques in the vorticity direction to the particles. At high volume fractions $\phi\geq 0.48$, where the ordering transition occurs, the suspension can be more ordered or disordered depending on the sign of the external torque. Applying negative torque can hinder the ordering transition, which is then accompanied by an increase in the shear viscosity. As a consequence, contrary to previous results at low volume fractions ($\phi\leq 0.20$) [6], the shear stress of the suspension is not reduced dramatically by negative torques. On the other hand, a positive torque has a favorable effect on the hexagonal order. However, above a certain threshold, the order begins to be weakened by the positive torque. At moderate volume fractions $\phi\leq 0.4$, the shear and vortex viscosities are neither sensitive to the sign nor the magnitude of torque.

The hydrodynamic interactions between particles are computed by the force-coupling method (FCM). In FCM, the long-range multibody interaction is fully resolved by solving the Stokes equations with regularized low-order multipoles [11]. FCM has been successfully applied for the analysis of MR flows [12]. Recently, a new modification of FCM to solve long-range multibody and viscous lubrication interactions simultaneously is presented [13] and used for concentrated suspensions in wall-bounded flows [10,14].

We focus on the volume fractions around which an ordering transition of non-Brownian suspensions occurs, $\phi = 0.48-0.52$ [10]. The computational domain is $H_x \times H_y \times H_z = 30a \times 20a \times 20a$, in which a is the particle radius and H_x, H_y, and H_z denote the lengths of the domain in the velocity (x), velocity-gradient (y), and vorticity (z) directions, respectively. Periodic boundary conditions are used in the horizontal directions (x and z). The computational domain is bounded by two parallel walls located at $y=0$ and H_y. The lower wall is fixed and the upper wall is moving in the x

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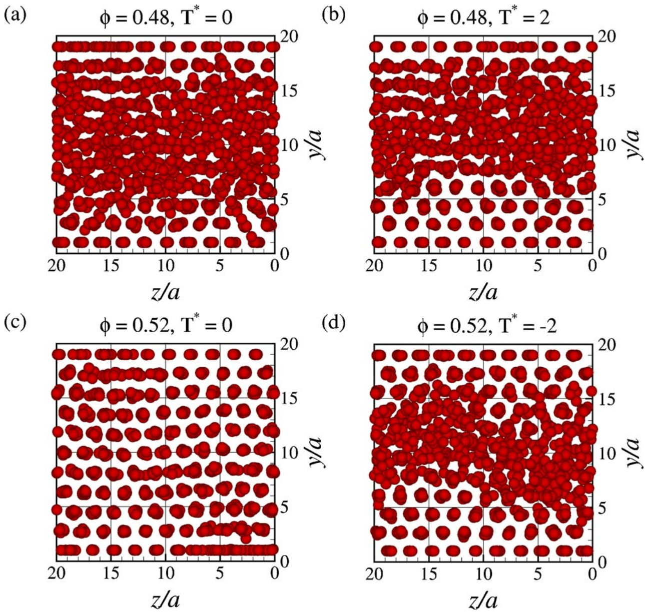

FIG. 1. (Color online) Snapshots (end view) for [(a) and (b)] ϕ=0.48 and [(c) and (d)] ϕ=0.52. For visualization, the particle radius is reduced to 1/2 of the actual size.

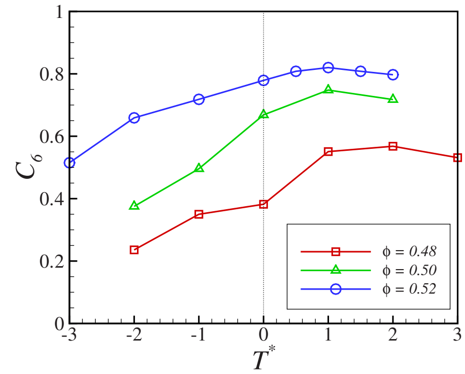

FIG. 2. (Color online) The order parameter C_6 as a function of the nondimensional torque T^*.

direction with velocity $V_{upp}=\dot{\gamma}H_y$, where $\dot{\gamma}$ is the nominal shear rate. The external torque is varied between $-3 \leq T^* \leq 3$, in which T^* is the torque normalized by the fluid viscosity μ_0 and $\dot{\gamma}$, $T^*=T/8\pi\mu_0\dot{\gamma}a^3$. In Couette flow, the suspended particles rotate in the clockwise (negative) direction. When a negative torque is applied, the particles rotate faster and a positive torque retards the rotation. To model irreversible forces, an elastic contact force is used when the shortest distance between two particle surfaces ($a\epsilon$) is smaller than $0.01a$ [14]. The magnitude of the contact force is set to keep the minimum separation distance $a\epsilon_{min}\simeq 0.002$ in all simulations.

Initial random configurations for the simulations are generated by a molecular dynamics procedure. First, the simulations with zero torque are allowed to evolve until the suspension reaches a stationary state. Then, an external torque is applied to the suspension. It usually takes about $t\dot{\gamma}\simeq 100$ to reach a new stationary state after a torque is applied.

Figure 1 shows illustrative examples of the effects of an external torque on ordering transitions. For $H_y/a=20$, an ordering transition begins around $\phi=0.48$ [10]. At $\phi=0.48$, the suspension is in a mixed disordered-ordered state whereby a hexagonal order exists near the wall with a disordered state in the core of the channel [Fig. 1(a)]. The suspension has a fully hexagonal order across the whole channel when $\phi \geq 0.52$ [Fig. 1(c)]. When the positive torque $T^*=2$ is applied to the suspension in a mixed state ($\phi=0.48$), the hexagonal order near the wall is more pronounced [Fig. 1(b)]. On the other hand, at $\phi=0.52$, the hexagonal order in the core of the channel is disturbed after the particles are subjected to the negative torque $T^*=-2$ [Fig. 1(d)].

To investigate the hexagonal order quantitatively, Kulkarni and Morris [8] suggested a hexagonal order parameter C_6,

$$C_6 = \left(\int_0^{2\pi} g(\psi)\cos(6\psi)d\psi \Bigg/ \int_0^{2\pi} g(\psi)d\psi \right), \qquad (1)$$

in which ψ denotes the azimuthal angle measured from the positive vorticity (z) direction and $g(\psi)$ is the pair-distribution function in y-z plane averaged over the radial interval $2a<r<2.1a$. $C_6=1$ for a perfect hexagonal order in y-z plane and $C_6=0$ if the suspension microstructure is isotropic. C_6 for different ϕ is shown in Fig. 2 as a function of T^*. It is clearly seen that the hexagonal order is always weakened by the negative torque. On the other hand, the effect of positive torque on the suspension is not straightforward. When a positive torque is applied, C_6 increases at first and then starts decreasing slowly after a threshold T^*_{crit}. T^*_{crit} seems to depend on the order state at $T^*=0$. Both for $\phi=0.50$ and 0.52, T^*_{crit} is observed around $T^*\simeq 1$, while T^*_{crit} for $\phi=0.48$ is found at larger T^*, $T^*\simeq 2$.

The shear stress of suspension flows at low Reynolds number in the presence of particle torques is [14,15]

$$\tau^* = 1 + \langle \sigma^*_{xy} \rangle + 3\phi T^*, \qquad (2)$$

in which $\langle \cdot \rangle$ denotes an average over the whole suspension, and τ^* and σ^*_{xy} are, respectively, the shear stress and x-y component of the symmetric part of the particle stress tensor normalized by $\mu_0\dot{\gamma}$. The sum of the first two terms on the right-hand side of Eq. (2) corresponds to the effective shear viscosity; $\mu_s/\mu_0=1+\langle\sigma^*_{xy}\rangle$. In suspensions under steady shear, normal relative motions between particles, which may occur during tumbling of a particle doublet by the shear flow, generate stresslets proportional to the inverse of the gap between particles ($\sim 1/\epsilon$), which is a major contributor to μ_s [16]. However, once the hexagonal order is developed, most particle interactions are tangential relative motions between particle strings, whose contribution to $\mu_s(\sim \log\epsilon)$ is much smaller than that from the normal motion. As a consequence, in the previous studies on suspensions under a steady shear, the decrease in μ_s is observed if a hexagonal order is present in suspensions [8–10].

The shear viscosity is shown in Fig. 3(a). In general, μ_s is a decreasing function of T^*. At $\phi=0.40$, where the suspension is in a disordered state, μ_s is much less sensitive to the external torque. There is only less than 10% difference be-

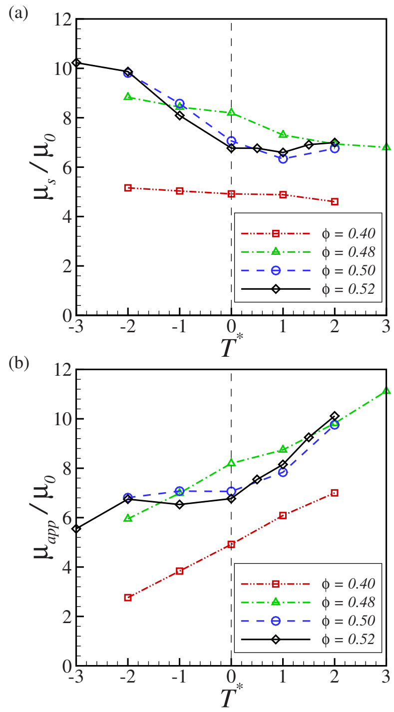

FIG. 3. (Color online) The (a) shear and (b) apparent viscosities as functions of the nondimensional torque.

tween μ_s for $T^*=-2$ and 2. At $\phi=0.48$, in which the ordered region is confined near the wall, the changes in μ_s by negative T^* is not significant similar to $\phi=0.40$. A dramatic increase in μ_s is observed at $\phi\geq 0.50$ when negative T^* is applied. As the hexagonal order is disturbed by negative T^*, a disordered region emerges in the channel core, which in turn results in the increase in μ_s. On contrary, the decrease in μ_s by positive T^* is most pronounced for $\phi=0.48$; μ_s is decreased about 15% by changing T^* from 0 to 2. However, when a positive torque is applied on the already ordered suspensions ($\phi\geq 0.52$), the changes in μ_s are not noticeable. Local minima of μ_s are observed for $\phi=0.50$ and 0.52, which correspond to the maxima of C_6 in Fig. 2.

Although the shear viscosity is an important parameter in studying suspension rheology, it is not easy to obtain μ_s directly in the experiments of sheared suspensions under an external torque. Here, we show the apparent viscosity, which can be obtained by measuring the shear stress on the top wall. The apparent viscosity of suspension μ_{app} is defined by a simple constitutive equation, $\tau=\mu_{app}\dot{\gamma}$, or $\mu_{app}=\mu_s+3\mu_0\phi T^*$. The definition of μ_{app} implies that, if suspension microstructure or μ_s is not altered by an external torque, μ_{app} can be increased or decreased by changing T^* and it may be possible to observe even a "negative viscosity" [6]. The theoretical prediction of the "negative ER effect" by [6] assumes that μ_s is a function of ϕ only, i.e., the microstructural change is negligible. The result for $\phi=0.40$ indicates that the assumption in [6] is indeed valid at lower volume fractions ($\phi\leq 0.40$). However, once a ordering transition occurs, μ_s becomes a function of both ϕ and T^* and the previous analysis is no longer applicable in this regime. If suspensions are in a ordered state, the magnitude of negative T^* needed to observe the negative ER effect would be much higher than that predicted by the analytical model [6].

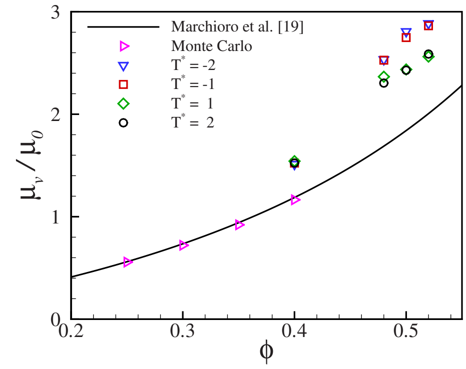

FIG. 4. (Color online) Variations in the vortex viscosity μ_v as a function of volume fraction for different values of the nondimensional torque in a confined shear flow. Also shown are results of Monte Carlo simulations for randomly seeded suspensions in a homogeneous uniform shear flow, compared to prior results [19].

The apparent viscosity is shown in Fig. 3(b). As μ_s is only weakly dependent on T^* at $\phi=0.40$, μ_{app} is seen to be a linear function of T^*. A notable change in μ_{app} is observed when $\phi\geq 0.50$. For $\phi\geq 0.50$, the contribution from negative T^* to μ_{app} is somehow balanced by the increase in μ_s. As a result, μ_{app} remains almost unchanged regardless of the magnitude of negative T^* in the range of the present study. For $\phi=0.52$, it is observed that, after a threshold $T^*\simeq -2$, μ_{app} begins to decrease.

One of the important rheological parameters in ER or MR fluids is the vortex viscosity μ_v, which is a measure of the hydrodynamic resistance to an applied external torque. The vortex viscosity μ_v is defined as [17,18]

$$\frac{\mu_v}{\mu_0}=\frac{3\phi T^*}{2\langle\Omega_z\rangle/\dot{\gamma}+1}, \tag{3}$$

in which $\langle\Omega_z\rangle$ is the averaged angular velocity of the particles in the vorticity direction. In an infinite domain, Marchioro *et al.* [19] calculated μ_v by a Monte Carlo procedure for a wide range of ϕ. For a comparison, μ_v computed by FCM in a triperiodic domain is shown in Fig. 4 together with the numerical fitting curve by [19]. μ_v in the present simulation is estimated from an ensemble average of 100 random configurations for each ϕ. The agreement with [19] is excellent. Due to the absence of any microstructure, μ_v obtained by the Monte Carlo approach does not depend on the sign or magnitude of T^*.

In Fig. 4, the vortex viscosity estimated by dynamic simulations of the confined steady shear flow is presented. At $\phi=0.4$, it is observed that μ_v obtained in the dynamic simulations is larger than that by the Monte Carlo approach in an infinite domain. In the previous boundary element computation of confined sheared suspensions, the vortex viscosity of confined suspensions was slightly lower than that of the infinite domain [18]. Hence, the increase in the vortex viscosity seen in the present dynamic simulations may be due to the formation of hydroclusters [20]. At $\phi=0.40$, μ_v is not sensitive to the signs and magnitudes of T^*. On the other hand, a bifurcation of μ_v is observed when $\phi\geq 0.48$, where the hexagonal order is present in the suspension. In general,

μ_v for negative T^* is always larger than that for positive T^*. At $\phi=0.48$, μ_v for positive T^* is a decreasing function of T^* for $T^* \le 2$, as the suspension becomes more ordered, while the effect of the negative torque, disturbing the order structure, is not noticeable. It seems that, because the suspension is already in a disordered state except near the wall, μ_v is less sensitive to the magnitude of torque for negative T^*. On the contrary, $\phi \ge 0.50$, μ_v does not change significantly when $T^* > 0$.

The changes in the ordered state correspond to the changes in the angular velocities of the suspended particles. In a homogeneous suspension, the additional hydrodynamic force from neighboring particles due to the external torques tend to be canceled out. In a wall-bounded sheared suspensions, however, the symmetry in the suspension microstructure is broken and the response of the suspension to an external torque now depends on the sign of the torque. The particle-wall resistance relation for the hydrodynamic torque in a shear flow is given by [14,21]

$$Y^C \Omega_z^D / \dot{\gamma} = T^* + 3Y^A (V_x - V_x^\infty)/4a\dot{\gamma} - Y^H = T^* + T_G^*, \quad (4)$$

where Ω_z^D is the retarded angular velocity $\Omega_z^D = \Omega_z + 0.5\dot{\gamma}$, Y^C, Y^A, and Y^H are the resistance functions, which depend on the distance from the wall, V_x is the translational velocity in the velocity direction, and V_x^∞ is the background velocity by the imposed shear rate. When a particle is close to the wall, both Y^C and T_G^* are positive, which indicates that the suspended particles near the wall rotate more slowly than the bulk when $T^*=0$. Similarly, for negative T^*, $|T^* + T_G^*| < |T^*|$ and the particles near the wall rotates slower than those in the core of the channel. On the other hand, if a positive torque is applied, $|T^* + T_G^*| > |T^*|$, indicating $|\Omega^D|$ for the particles near the wall becomes larger than that in the core. The asymmetric response to the sign of T^* may be explained by these wall-induced hydrodynamic interactions. As the dynamics of the wall layer is decoupled from that in the core before an ordering transition occurs, the wall effects are localized near the wall and the bulk dynamics does not change noticeably. However, near the ordering transition, a long-range correlation develops and the wall-induced hydrodynamic interactions may alter the overall suspension dynamics.

In summary, we find that ordering transitions in sheared non-Brownian suspensions confined by two parallel walls can be altered by applying an external torque in the vorticity direction on the particles. It is observed that negative torque always weakens the order structure, which in turn results in the increase in the shear viscosity. The dramatic decrease in the shear stress by the negative torque, which has been seen in lower volume fractions [6], is not observed once the hexagonal structure is present in suspensions. The hexagonal order is more pronounced with a positive torque. However, above a threshold, the order structure begins to be disturbed. Due to the changes in the order structure, the rheological parameters exhibit nonlinear responses to the applied torque. Here, we focus only on the suspensions in the channel whose height is $H_y/a=20$. However, as shown in [10], due to the complex interplay between the shear induced hydrodynamic interaction and the confinement effects, ordering transitions and rheological parameters show a complex behavior strongly dependent both on H_y/a and ϕ. Further investigations for a wide range of H_y, ϕ, and T^* are required to fully understand order structures and the effects of external torques in confined suspensions.

In this Brief Report, we have not specified a way to impose an external torque on the particles. The external torque may be achieved by applying magnetic or electric fields as seen in ferrofluid or ER fluid. However, in concentrated suspensions, electric or magnetic coupling between particles may be important, which is not considered in the present Brief Report. The effect of electric or magnetic coupling between particles is a subject of further investigation.

We thank Professor E. Climent for helpful discussions. An allocation of advanced computing resources is provided by the National Science Foundation under Grant No. TG-CTS090097. The computations were performed on Kraken (a Cray XT5) at the National Institute for Computational Sciences (http://www.nics.tennessee.edu/).

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