Local and Large-Scale Structure in Sheared Suspensions and Their Impact on Macroscopic Properties

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1 Introduction

Suspensions of Brownian solid particles dispersed in liquid have rheological behavior spanning the range from that of a Newtonian fluid at low particle volume fraction ϕ to, at large ϕ, a highly non-Newtonian fluid exhibiting shear thinning and thickening and normal stress differences (Stickell & Powell 2005), and eventually jamming at maximum packing, ϕ_m. At the same time that the rheology of the suspension changes, its microstructure undergoes a striking transition as well. Microstructure in suspensions generally refers to the pair distribution function, $g^{(2)}(\boldsymbol{r})$ or $g(\boldsymbol{r})$, which represents the probability of finding a pair of particles at a separation $\boldsymbol{r}$. As the shear rate increases, $g(\boldsymbol{r})$ changes from the equilibrium-like distribution in the absence of shear, dominated by excluded volume effects, to a pronounced accumulation of pair probability at contact at high shear, with accompanying regions of strong correlation in the shear plane around the axis of compression and, at high enough ϕ, near the x axis of the bulk flow $U_x = \dot{\gamma} y$, and a particle-deficient wake in the extensional zone (Lootens *et al.* 2004). Beginning with Bachelor & Green (1972), there have appeared a number of studies of the non-equilibrium pair structure of sheared suspensions. Echoing approaches of liquid state theory, where knowledge of pair distribution $g(\boldsymbol{r})$ allows prediction of thermodynamic functions for a system of particles subjected to pairwise potential, investigation of non-equilibrium suspensions have aimed to clarify the connection between macroscopic rheology and microstructure of suspension as a function of ϕ, Péclet number (Pe, ratio of shear to Brownian motion) and interparticle forces. Over the last several decades, investigators have proposed theoretical approaches to calculation of $g(\boldsymbol{r})$, mostly in the dilute limit, by employing the Smoluchowski advection-diffusion equation, subject to various simplifications and assumptions, for low and high Pe. Armed with the knowledge of pair-distribution, predictions then can be made of the influence it is expected to have on the suspension rheology and other properties. While the pair probability is the centerpiece of structural understanding, it cannot provide information on larger-scale structure. On the other hand, it has long been known that strongly sheared dense suspensions are prone to forming large clusters involving a great many particles, capable, e.g., of spanning the unit cell in a computer simulation (Bossis & Brady 1989).

In this study we investigate the triplet microstructure of a sheared suspension of Brownian particles. The following are some of the reasons why the triplets and their correlations are important:

1. The triplets provide a natural link from the pair level to many-particle arrangements (e.g., clusters) and therefore are expected to be important in the structural analysis of concentrated, strongly sheared suspensions.

2. A triplet's three particle centers always define a unique plane, unless they all happen to lie on the same line (a very special and important case that we will closely examine in this paper). Thus, in general, triplets can be thought of as planar objects and that property sets them apart from pairs which are strictly linear objects. Therefore, the triplets are expected to allow a wider array of measures and techniques for mapping out the spatial dependence of a richer spectrum of structures in the suspension.

3. It is known from liquid state theory that triplet correlations are important whenever many-body interactions (higher than the level of pairs) are expected to play a prominent role. As this is indeed the case with concentrated suspensions, we expect the proposed description at the triplet level to be very useful. One possibility is to describe force interactions among three particles at two levels: the so-called direct (particles 1 and 2, considered as a pair) and indirect (particle 3, regarded as an outside influence on the 1-2 pair and interacting with particle 1 via particle 2) force (Lionberger & Russel 1997).

4. In the Smoluchowski-type analysis of N-particle probability distribution, an integration to the pair level leads to an equation for the pair-distribution function in terms of the triplet distribution, giving rise to a well-known closure problem (Lionberger & Russel 1997).

5. In liquid state theory, the triplet distribution function plays an important role in phase transitions and in the critical region (Buff & Brout 1960, Dhont & Verduin 1997).

Bildstein & Kahl (1994) studied triplets in equilibrium hard-sphere systems simulated by molecular dynamics at a variety of packing fractions and reported non-trivial structures of the so-called isosceles triplets (where two of the three vectors making up the triplet are of equal length); they used the data from their simulations to discuss the merits of different theoretical approaches to the direct calculation of triplet correlation. However, while the triplet-distribution function has appeared in theoretical work treating non-equilibrium sheared suspensions of Brownian particles (Lionberger & Russel 1997), to our knowledge, no actual triplet structure results have been reported to date.

It is the goal of this work to present for the first time an investigation of the triplet structure of monodisperse suspensions of Brownian spherical particles in shear flow at zero Reynolds number (Stokes flow) and to demonstrate the ways in which the analysis of the triplet structure can motivate a further investigation into clusters and stress chains (the term we use to denote stress bearing linear clusters ubiquitous, as we shall see, in concentrated strongly sheared suspensions). We employ conventional (Brady & Bossis 1988; Phung, Brady & Bossis 1996) and accelerated Stokesian Dynamics simulations (Banchio & Brady 2003) to generate realizations of periodically replicated two- and three-dimensional unit cells of Brownian particles in shear flow, spanning the range of Pe from near equilibrium (Pe=0.01) to conditions of very strong shear (Pe=1000). We generate and track distributions of triplet shapes in and out of the plane of shear and correlate them to the simultaneously occurring changes in the particles' pair correlation. The outline of this paper is as follows. We present definitions of pair and triplet probabilities in Section 2; the specifics of the model system and the simulation method are described in Section 3; Section 4 presents the results of the isosceles triplet structure study and, in Section 5, we conclude with a demonstration of preliminary results of cluster and Voronoi cell analyses of the suspension and suggest ideas for future work.

2 Pair and triplet probability distribution functions

In a suspension of particles, the pair-distribution function, $g^{(2)}$ or g, is the normalized probability of finding a second particle at $\boldsymbol{r}$, given one at the origin,

$$P_{1|1}(\boldsymbol{r}|0) = ng(\boldsymbol{r}).$$

At equilibrium, $g(\boldsymbol{r}) = g(r)$, *i.e.* there is spherical symmetry. Pair correlation in equilibrium HS colloids is therefore identical to that of HS gas molecules, and depends only upon packing fraction; static structure and thermodynamics have a one-to-one correspondence with those in the hard-sphere gas (Russel *et al.* 1989). The pair distribution is experimentally accessible from light scattering experiments (Bender & Wagner 1995; Newstein *et al.* 1999), where it is measured as the FT($g(\boldsymbol{r})$) or the structure factor $S(\boldsymbol{k})$. In sheared systems, the structure can be probed also by direct microscopy (Parsi & Gadala-Maria 1987), recently with confocal equipment (Semwogerere & Weeks 2005). Experiments (Parsi & Gadala-Maria 1987) and simulations of non-equilibrium systems (Phung *et al.* 1996; Morris & Katyal 2002; Stickel & Powell 2005) show that strong shear causes extreme angular distortion of $g^{(2)}$, qualitatively altering the rheology.

The probability, given a particle at the origin, of simultaneously finding a second particle at $\boldsymbol{r}$ and a third at $\boldsymbol{s}$ is

$$P_{2|1}(\boldsymbol{r}, \boldsymbol{s}) = n^2 g^3(\boldsymbol{r}, \boldsymbol{s}),$$

where $g^{(3)}$ is the *triplet* distribution function; $g^{(4)}$, $g^{(5)}$, ... follow in similar fashion. The need for these functions arises when one attempts to obtain a direct solution for $g^{(2)}$, leading to a hierarchy relating $g^{(2)}$ to $g^{(3)}$, $g^{(3)}$ to $g^{(4)}$, and so forth. This hierarchy can be broken most simply by the Kirkwood superposition approximation (KSA) in which $g^{(3)}$ is taken as the product of the three (independent) pair distributions, or the quite successful hypernetted chain equation and Percus-Yevick approximation. Sampled $g^{(3)}$ from simulation of atomic liquids and hard sphere systems (Muller & Gubbins 1993; Bildstein & Kahl 1994), shows KSA varies widely in its accuracy near contact. Following Bildstein & Kahl (1994), we restrict our attention in this work to isosceles configurations at contact, i.e. we will map out the function $g^3(2, 2, \varphi)$, where φ (or ϑ in Bildstein & Kahl's notation) is the angle formed by the equal-length sides of the isosceles triplet and the separation r, normalized by the particle radius a is equal to 2 at contact. Two isosceles configurations are of special interest. They are the limiting cases of the equilateral triangle, where all three particles are in contact with each other—we term this arrangement the "closed" triplet; and the fully "open" triplet, where the two equal pairs make a 180-degree angle. Bildstein & Kahl (1994) saw an accumulation of probability at $\varphi = \pi/3$, indicating that the closed triplet is the favored configuration at equilibrium conditions. In the results section of this paper, we confirm that triplets in near equilibrium suspensions obey the distribution obtained by Bildstein & Kahl and investigate how the prevalent triplet configuration changes as a function of the Pe number.

3 Model system and simulation method

System characterization: At equilibrium, a monodisperse hard-sphere (HS) suspension of Brownian particles of radius a is characterized by the particle volume fraction, $\phi = 4\pi n a^3/3$ (in monolayer, areal fraction ϕ_A), where n is the number density. In the absence of interparticle forces, thermal energy kT serves only as a scaling factor that sets rates of particle motion and the osmotic pressure. Shear flow introduces another time scale, and the dimensionless Péclet number, $Pe = 6\pi\eta\dot{\gamma}a^3/kT$ characterizes the relative time scales for Brownian motion to shear flow; here $\dot{\gamma}$ is the shear rate and η is the fluid viscosity.

Simulation method: Stokesian Dynamics (SD) has been in development for over 20 years and its detailed description can be found in Brady & Bossis (1988), Brady *et al.* (1988) and Phung *et al.* (1996), while accelerated Stokesian Dynamics (ASD) is described in Sierou & Brady (2001) and Banchio & Brady (2003). In SD simulation of N Brownian particles suspended in Stokes flow, particle trajectories are obtained by solving

$$0 = \boldsymbol{F}^H + \boldsymbol{F}^B + \boldsymbol{F}^P \tag{1}$$

where $\boldsymbol{F}^H = -\boldsymbol{R}\cdot(\boldsymbol{u} - \boldsymbol{U}^\infty)$ is the N-particle vector of hydrodynamic forces-plus-torques (size $6N$ in 3D) given in terms of a configuration-dependent resistance tensor $\boldsymbol{R}$ and the deviation of the particle kinematics (velocities and velocity gradient in $\boldsymbol{u}$) from the imposed flow, $\boldsymbol{U}^\infty$, here simple shear ($U_x^\infty \sim y$); $\boldsymbol{F}^B$ is the Brownian force satisfying fluctuation-dissipation at equilibrium, and $\boldsymbol{F}^P$ represents interparticle forces which may be written as the gradient of pairwise potentials. Vanishing total force and torque is a requirement of no inertia.

Calculation of the motion requires assembling a "grand resistance tensor" describing the force and its first surface moment (antisymmetric = torque, $\boldsymbol{T}$; symmetric = stresslet, $\boldsymbol{S}$) to the particle motions (in particular the deviation $\boldsymbol{u} - \boldsymbol{U}$) and thus the solution for the motion allows calculation of the rheological properties through the stresslet (likewise composed as $\boldsymbol{S} = \boldsymbol{S}^H + \boldsymbol{S}^B + \boldsymbol{S}^P$ and thus assignable to the generating mechanism) simultaneous with locations/velocities, critical for structure-property relations. The method captures near- and far-field physics and compares well to experiment (see Phung *et al.* 1996; Morris & Brady 1998).

Recently, Sierou & Brady (2001) described ASD, in which far-field mobility matrix computation and its inversion (operations $O(N^2)$ and $O(N^3)$, respectively) were replaced using particle-mesh-Ewald techniques with operations of $O(N \ln N)$ permitting larger N; ASD was subsequently augmented to include Brownian motion (Banchio & Brady 2003). We utilize both SD and ASD, beginning with SD of monolayers for speed and to facilitate analyses and visualization.

Cases simulated: We have simulated two-dimensional cells with monolayers of 124 particles at areal fractions ϕ_A=0.5 and 0.7. The three-dimensional simulation was completed using ASD of 999 particles in the unit cell at ϕ=0.467, corresponding to ϕ_A=0.7. The range of Pe studied is from 0.01 to 1000.

4 Results

Before describing the novel data on the triplet structure, we briefly review some of the results obtained by Morris & Katyal (2002) for the pair distribution function in concentrated strongly sheared suspensions (shown in Figure 1). Known features of $g(\boldsymbol{r})$ under shear are that it becomes elevated with extreme radial gradient near contact and strong anisotropy. Configurations having near-contact pair separation vector in the compressional quadrant are greatly favored.

In reporting the triplet structure data obtained in this study, we consider only contacting triplets, which form isosceles triangles with two sides of length the particle diameter. Two kinds of triplet selection procedures was used in order to obtain data on triplet distributions: in one procedure, we accounted for all possible triplets regardless of their orientation relative to the plane of shear (in other words, irrespective of the angle θ between the z axis—direction of vorticity—and the normal to the plane in which the triplet lies; in the other procedure, only the triplets lying close to the shear plane were allowed (in this case, the pairs making up the triplet are not allowed to make more than a 10^o angle with the shear plane). Figure 3 shows that triplet probability depends upon the angle between contacting pair vectors, in a manner strongly dependent upon Pe. At $Pe \ll 1$, the results are similar to the equilibrium results of Bildstein & Kahl (1994), obtained for hard spheres at $\phi = 0.45$, plotted in Figure 2. As Pe increases, the dominant close triplet moves progressively from an equilateral triangle (illustrated in Figure 3) toward a linear configuration – pronounced anisotropy in $g(\boldsymbol{r})$ carries over to the triplet. We report

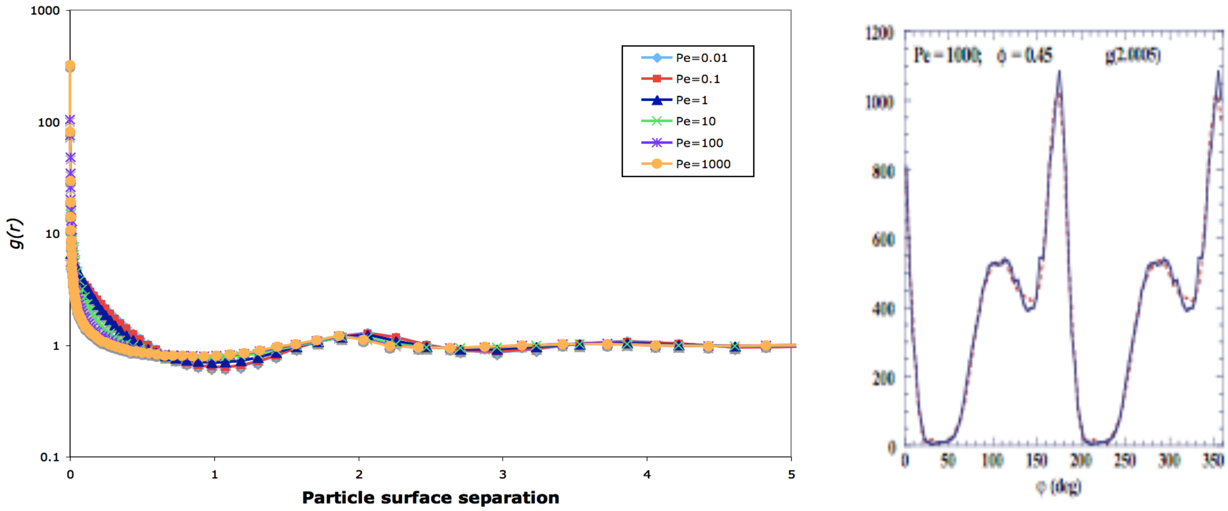

Figure 1: Left: angular average of $g(\boldsymbol{r})$ for 3D suspensions of $\phi = 0.467$ at $Pe = 0.1$ to 1000; right: shear plane contact (smallest sampled separation) $g(\boldsymbol{r})$ with angle measured relative to $+x$ axis in flow $\boldsymbol{U}_x^\infty = \dot{\gamma} y$ for $\phi = 0.45$ and $Pe = 1000$, adapted from Morris & Katyal (2002).

the results averaged with respect to the angle describing the orientation with respect to the flow, but the preferred angle depends upon ϕ; again, certain features of $g^{(3)}$ are readily understood in terms of $g(\boldsymbol{r})$. For nearly-contacting particles, this suggests a route to modeling the distribution of many-particle (> 3) clusters. In Figure 4 we present the distributions of orientations of both closed and open isosceles contacting triplets as a function of the Péclet number. We note that as Pe increases from 1 to 10, the distribution of closed triplets living in the shear plane is dramatically reduced and this explains the disappearance of the peak at 60^o in the plot on the right-hand side of Figure 3, which accounts for triplets residing in the shear plane. The distribution of the open triplets apparently undergoes changes at least twice as shear is increased: first, going from $Pe = 1$ to 10, there appears to be an shift in the distribution in favor of open triplets residing in the shear plane, which again helps to explain the simultaneously occurring dramatic change in the curves of Figure 3. However, as shear is increased further, the distribution of open triplets seems to tend to return to an equilibrium-like flat profile previously seen in low-shear systems. We defer to future work a thorough investigation of the details of this transition.

Finally, in Figure 5, we show the distribution of orientation of the open triplets in the shear plane, for $Pe = 1000$. This distribution looks remarkably similar to the picture of the anisotropy of the pair-distribution function reported by Morris & Katyal (2002)—cf. Figure 1 where we have included their results. We see here strong evidence of the anisotropy of $g^{(2)}$ being the driving force behind the formation of the open triplets—a favored feature in highly sheared suspensions (especially in the shear plane).

5 Conclusions

We have presented results of an investigation into the triplet structure of a concentrated sheared suspension of Brownian particles simulated by Stokesian Dynamics. Looking at the isosceles contacting triplet configurations, we have uncovered intriguing structural trends as a function of the Péclet number. While at low shear the distribution of the triplets was found to be very close to that found in an equilibrium hard-sphere gas, a very prominent change occurred between $Pe = 1$ and 10: the domination of equilateral, closed, triplets gave way to a prevalence of the open, chain-like configuration. The change was found to be even more pronounced when attention was restricted to triplets lying in the shear plane, where the open triplets' preferred orientation appears to mimic the anisotropy of the pair-distribution function. Also, the above-mentioned features are the most pronounced at $Pe = 10$; in systems subject to higher shear, at $Pe = 100$ and 1000, the features tend to attenuate somewhat, with at least one of them,

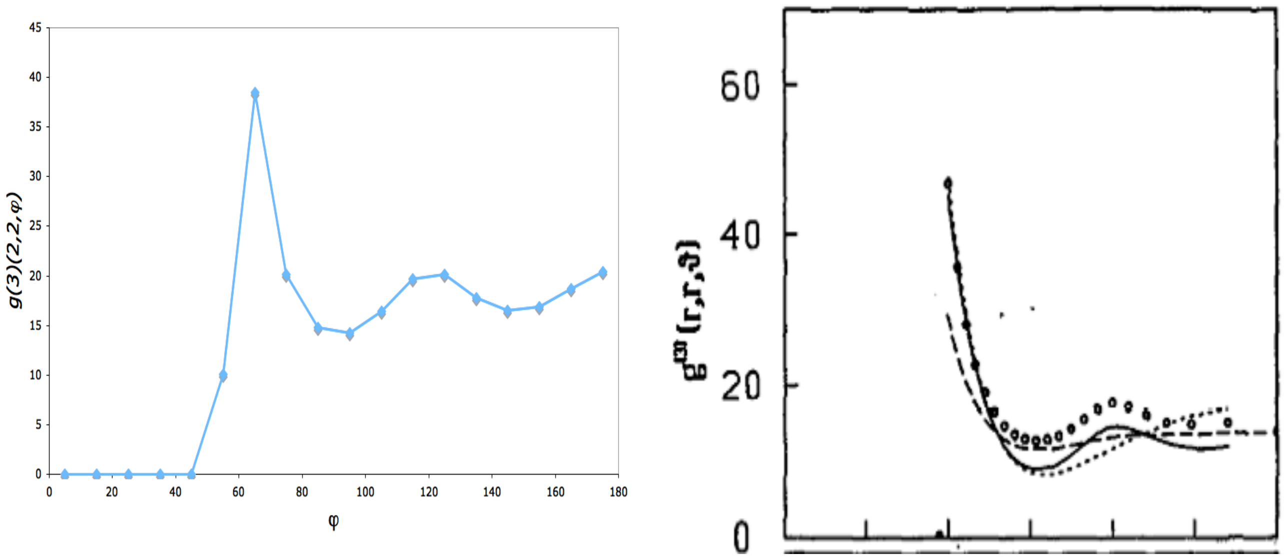

Figure 2: Left: triplet distribution of isosceles contacting configurations as function of angle between $\boldsymbol{r}$ and $\boldsymbol{s}$ from 3D simulations at $Pe = 0.01$ and $\phi = 0.467$, average over all available triplets. Right: triplet distribution of isosceles contacting configurations of hard spheres at $\phi = 0.45$, from Bildstein & Kahl (1994)—simulation results show as open circles, while the angle ϑ runs from 0 to 180^o in 30-degree increments.

a preferred orientation of open triplets in the shear plane, completely disappearing.

In future work, we intend to continue our investigation into the triplet structure of non-equilibrium suspensions with an emphasis on its influence on macroscopic properties. On the other hand, the findings of this study suggest a way of conducting an investigation of the issue of larger particle clusters. Below we present some preliminary ideas and results of an application of a cluster finding algorithm and a Voronoi tesselation algorithm for the purposes of identifying extended linear structure of contacting particles and their influence on the suspension rheology.

5.1 Clusters

Crucial to cluster modeling is data on clusters from a range of conditions. We have used the connectivity/cluster finding algorithm of Sevick *et al.* (1988), taking account of difficulties with spanning clusters in periodic simulations noted by Lee & Torquato 1988) for monolayers. For Brownian HS, defining contact for a pair below a minimum surface separation (in Figure 5, 10^{-3} radii), at $Pe > 10$ the tendency to clustering escalates rapidly. Results for the 3D and monolayer of $\phi_A = 0.7$ are suspect because spanning clusters become the norm in these simulations. Care must be taken to eliminate the spurious influence of these, and the point of these results is to illustrate how rapidly the condition is approached for small N (here 120).

5.2 Voronoi cell distributions

In two dimensions, a Voronoi diagram is the result of partitioning of a plane containing n points into n 'cells': the cells are polygons each containing exactly one point, with any position in the cell closer to that point than the other $n - 1$. In 3D, the cells are polyhedra. Here the points are the particle centers in sheared suspensions. In figures 7 and 8, Voronoi polygons in a monolayer are colored according to the level of the shear stress contribution of the particle in the cell (divided by cell area) at $Pe = 1$ and $Pe = 1000$, respectively; each figure shows the highest and lowest viscosity realization from an extended simulation with 120 particles. High-viscosity fluctuations are

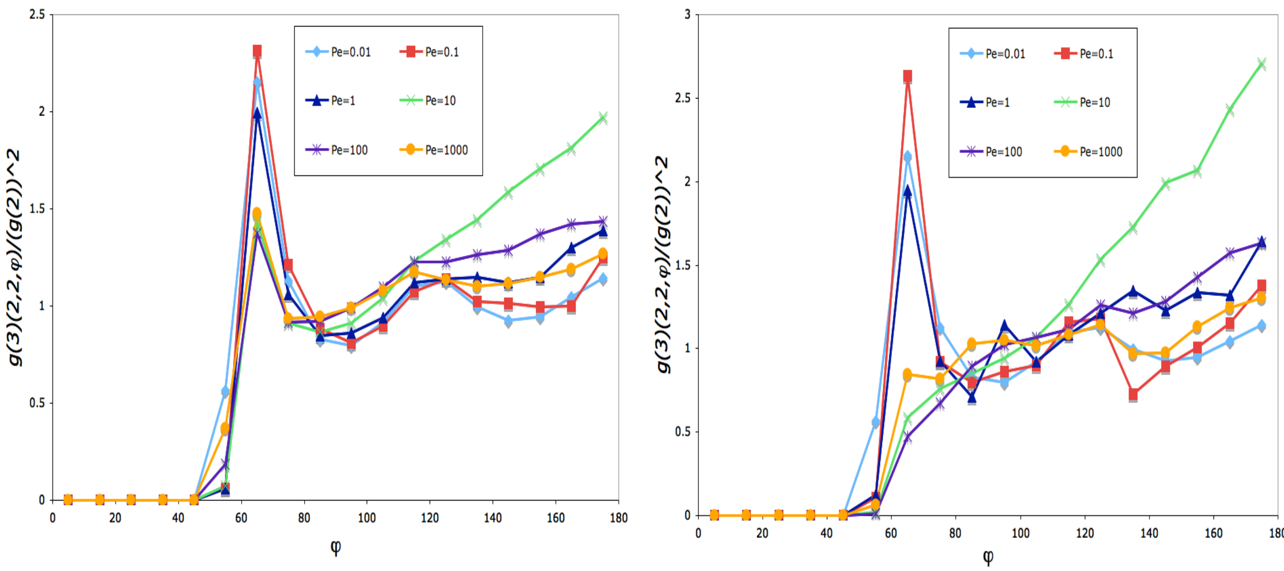

Figure 3: Left: triplet distribution of isosceles contacting configurations as function of angle between $\boldsymbol{r}$ and $\boldsymbol{s}$, normalized to $(g(2))^2$, from 3D simulations at $\phi = 0.467$, average over all available triplets. Near equilibrium (lowest $Pe = 0.01, 0.1$, and 1), the closed triplet configuration is preferred; at $Pe = 10, 100$, and 1000, open triplets are prevalent. Right: triplet distribution of isosceles contacting configurations in or near the plane of shear ($\theta = \pm 10^o$). For $Pe > 1$, the shoulder at $\Delta\varphi = 60^o$ completely disappears, suggesting the relative scarcity of closed triplets in the shear plane.

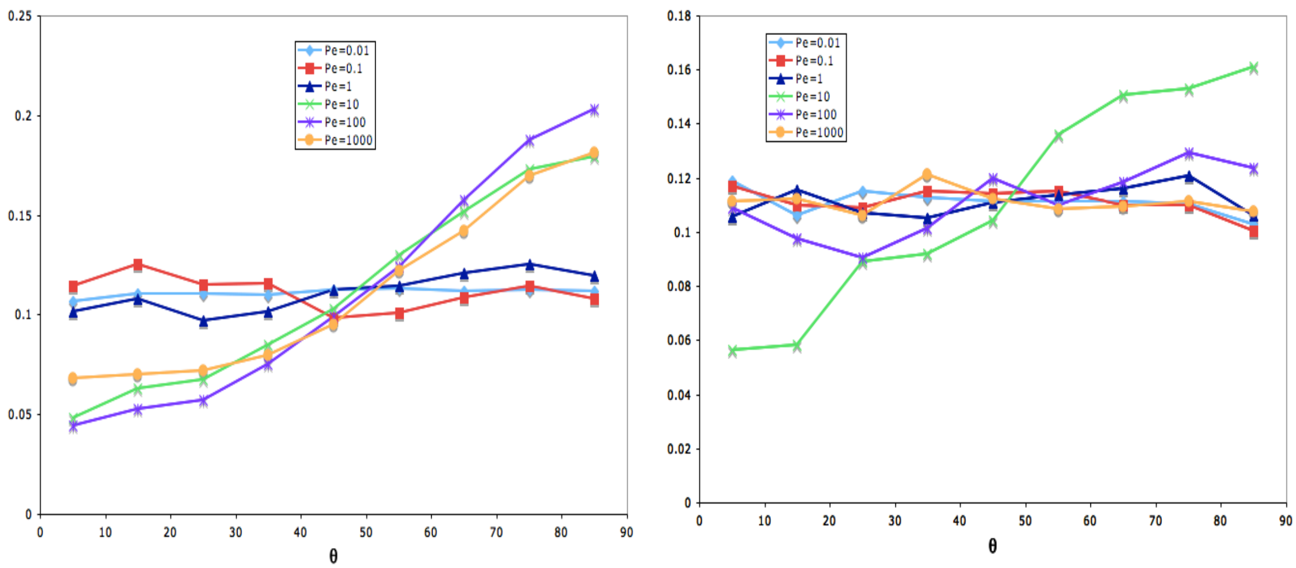

Figure 4: Left: distribution of equilateral closed contacting triplets as a function of the angle θ between the direction of vorticity (the z axis) and the normal to the plane of the triplet. Low values of θ correspond to triplets lying in or near the plane of shear, while high θ values (near $\pi/2$) represent triplets normal to the shear plane. The change in the distribution occurring between $Pe = 1$ and 10 supports the interpretation of the data shown in the previous figure. Right: distribution of isosceles open (to 180^o) triplets as a function of the angle θ between the z axis and the vector connecting particles 2 and 3 of the triplet.

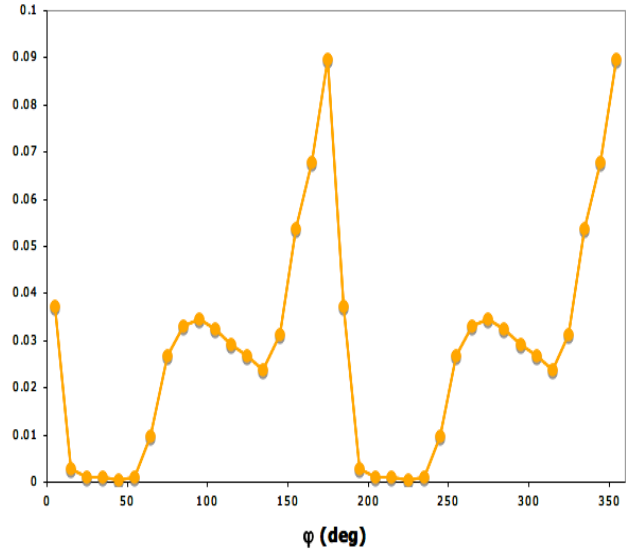

Figure 5: Distribution, at $Pe = 1000$, of open contacting isosceles triplets in the shear plane as a function of the angle φ between the direction of flow (the x axis) and the vector connecting particles 2 and 3 of the triplet. Compare this to the plot on the right-hand side of Figure 1, showing the anisotropy of the pair-distribution function at contact in the shear plane.

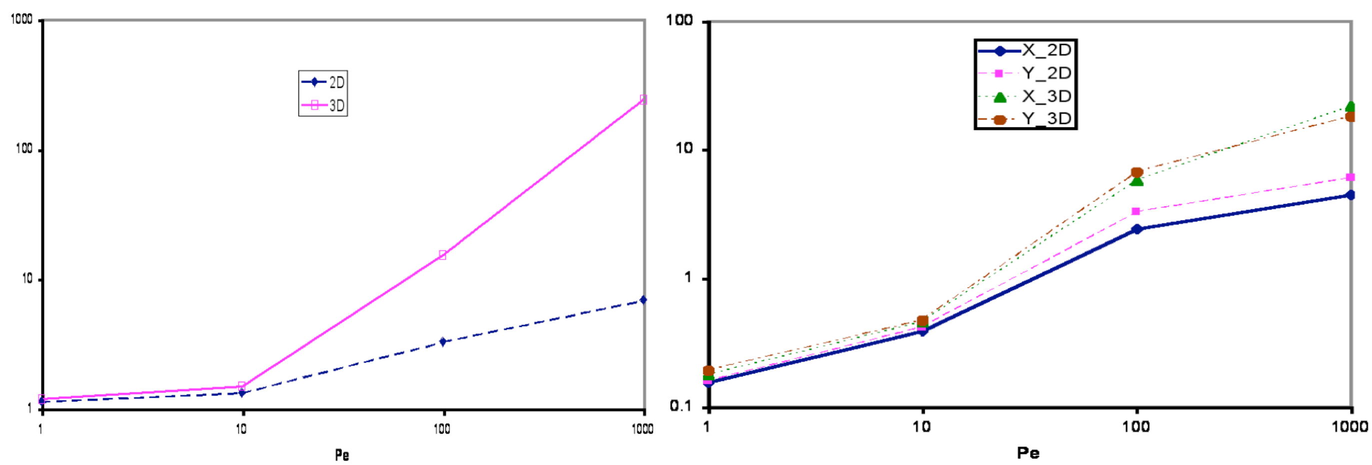

Figure 6: Cluster measures in monolayer (2D, $\phi_A = 0.5$) and fully 3D ($\phi = 0.33$) simulations. At left is the mean number of particles in a cluster and at right the linear extent in flow and gradient directions, as a function of Pe. The 3D results become ambiguous due to percolation across the unit cell above $Pe = 10$.

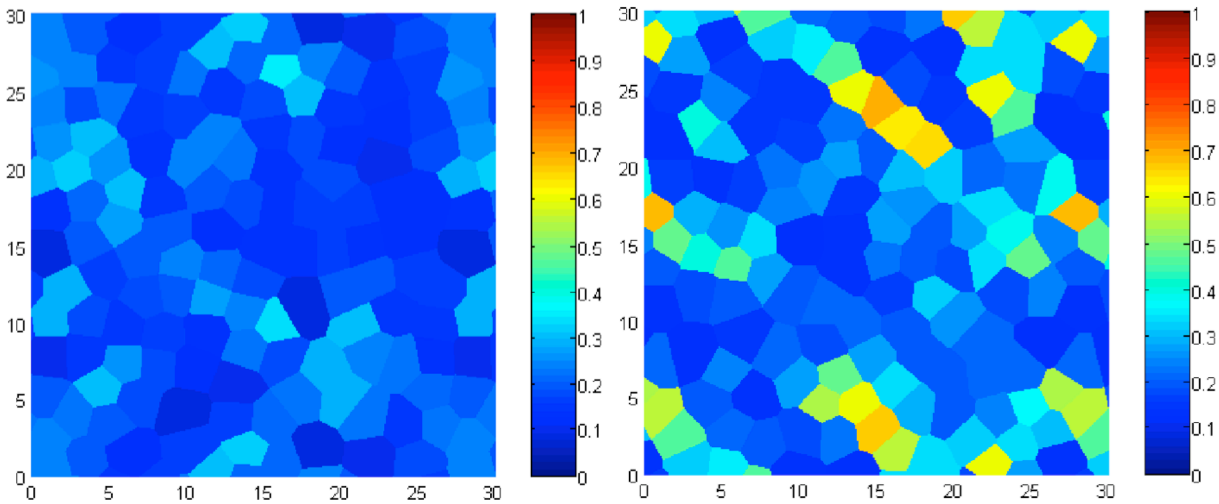

Figure 7: Particle Voronoi polygons and stress distribution at $\phi_A = 0.5$ and $Pe = 1$ in low (left, 85% of full simulation run average) and high (right, 121%) viscosity realizations of a 120-particle monolayer suspension (left and right, respectively). Polygons are colored according to the magnitude of the x-y (shear) component of the individual particle stresslet divided by that polygon area. Flow is to right at top, to left at bottom.

associated with structures extending along the compressional axis of the shear flow, with much larger structures observed at large Pe. We propose the term "stress chains" for these structures to denote the structure-rheology relationship they appear to embody.

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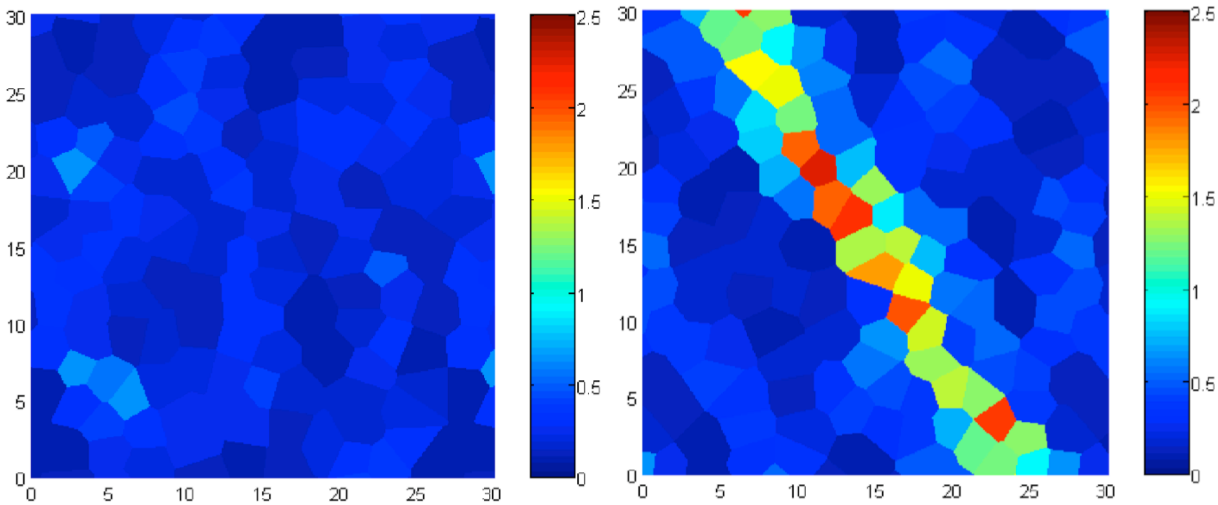

Figure 8: As in Figure 7, except $Pe = 1000$. Realizations are at low (left, 58% of full simulation run average) and high (right, 149%) viscosity.

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