Aggregation behavior of two spheres falling through an aging fluid

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We study the behavior of two spheres that settle along their line of center in a yield stress fluid that "ages:" a Laponite suspension. In such a fluid, the fluid flow behind a falling single particle can either exhibit a negative wake, i.e., an upward motion, or not, according to the stress exerted by the particle on the fluid. We show that, if their initial separation distance is smaller than 15 radii, two identical particles cluster whatever the wake's structure. In addition, in the conditions within which a negative wake is observed, we evidence an unexpected lateral motion of the spheres.

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The velocity field induced by a sphere settling in a less dense Newtonian fluid can be obtained analytically by solving the Stokes' equation provided the fluid is unbounded and the Reynolds number is small. Owing to the linearity of the Stokes' equation, the motion of two identical spheres can be inferred from the obtained velocity field [1]. As a consequence of the symmetries of the velocity field, two identical spherical particles interact in a symmetrical way and the doublet is stable whatever its configuration. The separation distance between spheres is thus expected to be constant, which is well verified experimentally provided the settling tank dimensions are large compared to the sphere's radius.

In complex fluids, i.e., fluids that exhibit nonconstant viscosities and/or elasticity, different behaviors have been observed [2,3]. According to the fluid's rheology and to their initial separation distance, two identical spheres sedimenting along their line of centers have been found to either form a vertical aggregate or remain at constant separation distance or even repel each other, the trailing sphere sedimenting more slowly than the leading one. These unexpected phenomena—that all occur at small Reynolds numbers at which inertial effects are negligible—have aroused interest in the past decade and both numerical and experimental efforts have been directed toward their understanding; the case of single spheres has in particular been widely studied in order to characterize and predict the velocity field within the fluid [4] and possibly infer information on the two-particle interactions. The absence of a stable separation distance between two particles in some polymer solutions has been related to the loss of fore-aft symmetry in the velocity field induced by the settling of a single particle. As a manifestation of this asymmetry, the existence of an upward motion of the fluid in a single particle's wake has in particular been widely documented. This so called "negative wake" has been observed in different complex fluids [5] and, although its physical origin remains unclear, the extensional rheology of the fluids is doubtlessly involved. The presence of such a negative wake has been invoked to retard the trailing spheres located beyond the stagnation point (where the flow reverses) and thus to lead to a "repulsion" between the two particles [4]; following this argument, spheres at smaller separation distances would, on the contrary, be entrained by the negative wake and further form an aggregated doublet. This analysis indeed relies on the properties of additivity that are valid in a Newtonian Stokes' flow. Their validity in complex fluids remains, however, unclear since, for instance, bubble coalescence (resulting from an attraction between bubbles) occurs in polymeric fluids in which negative wakes are also observed [6].

We present herein a study of the behavior of two spheres settling initially along their line of centers in a Laponite suspension. Laponite is a synthetic colloidal clay whose suspensions are known to exhibit yield stress, shear thinning, and aging i.e., an irreversible evolution of their rheological properties with time when at rest [7]. These suspensions constitute fluids of interest for two-particle studies since the velocity field around a single particle can either exhibit a negative wake or not, according to both the aging time and the stress exerted by the particle [8]. As this stress scales as $\Delta\rho$, the difference in density between the fluid and particles, the use of particles of different weights allowed us to study the influence of the wake's structure on two-particle behavior. We show in this paper that, at initial separation distances smaller than 15 particle radii, spheres sedimenting in a Laponite suspension form aggregates whatever the wake structure, in particular even in the conditions within which a negative wake is observed. The two-particle behavior is therefore a consequence of the modification of the fluid's state, itself resulting from the passage of the leading particle, and cannot be directly related to the velocity field around a single particle.

We have used calibrated macroscopic spheres of radius $a=0.75$ mm and of two different densities (aluminum of density 2.7 g/cm^3 and brass of density 8.7 g/cm^3). For each sphere's material a mass variation between spheres smaller than 2% has been found. The fluid is an aqueous suspension of Laponite (Rockwood) of concentration 2.5% w/w and of pH 10. The samples are strongly mixed and kept under a nitrogen atmosphere to obtained controlled physicochemical conditions. Prior to the settling experiments, the suspension is stirred for at least 30 min in order to produce a completely unstructured initial state. The fluid is further poured in the settling cell and left at rest for a time t_a during which it ages i.e., in particular its apparent yield stress increases. Since at a given aging time the velocities of aluminum and brass spheres strongly differ, different values of the aging times had to be chosen for the introduction of the aluminum and

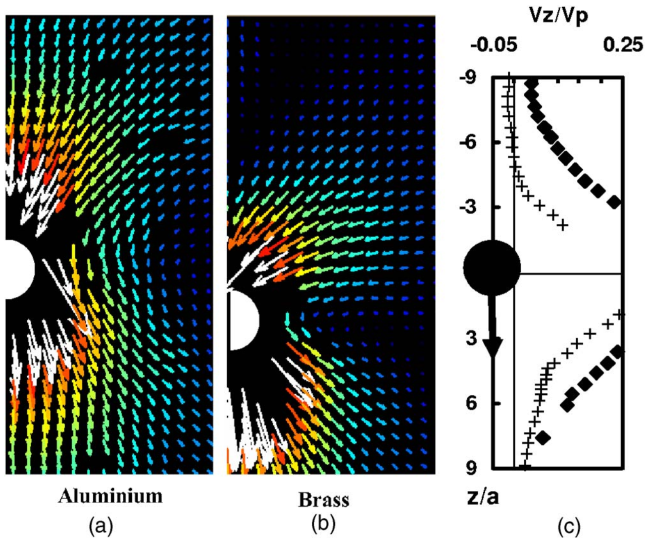

FIG. 1. (Color online) Velocity fields measured in the Laponite suspension for (a) the lighter (aluminum) and (b) the heavier (brass) spheres. The corresponding variations of the vertical component of the fluid velocity as a function of the vertical position, z from the aluminum (full diamonds) and brass (crosses) spheres are shown in (c). The fluid aging times are respectively $t_a=45$ min and $t_a=60$ min. The vector scales are different in (a) and (b) since the particle velocities differ by an order of magnitude.

brass particles in the cell i.e., respectively $t_a=45$ min and $t_a=60$ min. The settling velocities of single spheres are respectively $V_p \approx 0.4$ mm/s for the lighter (aluminum) particle and $V_p \approx 4$ mm/s for the heavier (brass) particle. In both cases the corresponding Reynolds number is smaller than 10^{-2}, the inertial effects are therefore expected to be negligible.

The fluid velocity around single particles has been measured using the Particle Image Velocimetry (PIV) technique which experimental details are given elsewhere [8]. Figure 1 displays the velocity fields thus obtained around the lighter (aluminum) particle and heavier (brass) particle. In both cases the flow exhibits no fore-aft symmetry: along the lighter sphere's centerline, the fluid vertical velocity v_z decreases monotonously away from the sphere in both directions but it decreases faster downstream than upstream; in the heavier particle case, the flow is confined closer to the particle and in the wake of the particle, beyond a stagnation point located at about five particle radii, v_z becomes negative. It further reaches a minimum after which it tends towards zero (not shown). The recirculating zones on each side of the spheres are a consequence of the finite size of the settling cell (whose smallest dimension is 2.4 cm) and are also observed in a Newtonian fluid. The same qualitative features have been observed for both spheres at aging times ranging from 30 min to 220 min [8], the behaviors described in the following are therefore expected to be observed in this range of aging times provided the Reynolds number remains small and the particles are in motion.

For the two particle experiments, the settling cell is cylindrical and made of Plexiglas; its inner diameter is 2.4 cm and useful height 17 cm. It is placed in a thermostatic bath with the same refraction index. The motions of the spheres are imaged using two CCD cameras whose optical axes are perpendicular in order to measure the vertical position z (the z axis being directed downward) as well as the radial position r of each sphere. The adopted magnification is a compromise between loss of precision on the radial motion and the need for the imaging over the largest vertical distance. The uncertainty upon the radial position of each sphere is $\pm\frac{a}{4}$. The spheres are released through a drilled cap which ensures that their centers are separated by a radial distance $r \leqslant 2a$. Only vertical center to center separation distances δz smaller than 15 sphere radii have been considered in order to perform accurate measurements on the radial positions. Note that we do not control the vertical plane within which the spheres fall, which renders impossible PIV measurements in the two-sphere experiments. The images are further digitized using commercial imaging software (Image Pro +, Media Cybernetics). Experiments performed in a Newtonian fluid (pure glycerin) showed the expected behavior, i.e., spheres falling at equal velocity and therefore maintaining a constant relative separation.

We have studied the motion of doublets of identical particles with different initial separation distances. Figure 2 shows the aggregation behavior of the doublets of respectively the lighter and heavier spheres at successive times. The pictures were taken along a fixed vertical line in the laboratory's frame. In both cases, the spheres that initially fall along their centerline within the experimental precision eventually form a stable vertical cluster. The presence of a negative wake therefore does not prevent the aggregation of the spheres at separation distances beyond the stagnation point. In addition, we have observed an unexpected radial motion of the heavier spheres: the leading sphere moves laterally (to the right in Fig. 2) prior to cluster formation. The relative motions of the trailing spheres are detailed in Fig. 3 that shows the spheres' trajectories in the frame of the leading spheres. In this frame and in the case of the doublet of the lighter particles, the radial velocity of the trailing sphere remains negative since its radial position monotonously decreases to zero within the experimental uncertainty. In the case of the doublet of the heavier particles, the motion of the trailing sphere is more complex: the radial distance between spheres first increases from its initial value up to 1.5 particle radius and further decreases to zero.

In a Newtonian fluid and within the point force approximation, the velocity $\vec{V}_i$ of each sphere is given at the first order by adding to $\vec{V}_S$, the Stokes' velocity, the fluid velocity induced in its center O_i by the fall of the other sphere, $\vec{v}_j(O_i)$, which yields $\vec{V}_i=\vec{V}_S+\vec{v}_j(O_i)$. In the Laponite suspension, the radial component of the fluid velocity around a single sphere is too small to be accurately measured but one can compare the vertical components of the particle velocities with the ones computed by analogy with the ones in a Newtonian fluid. Figure 4 shows the z component of the fluid velocity as a function of the radial distance and at given vertical distances z/a from the center of a single particle. For both brass and aluminum spheres the values of v_z are larger upstream of the particle than downstream. This asymmetry is a consequence of the time dependent properties of the fluid. By anal-

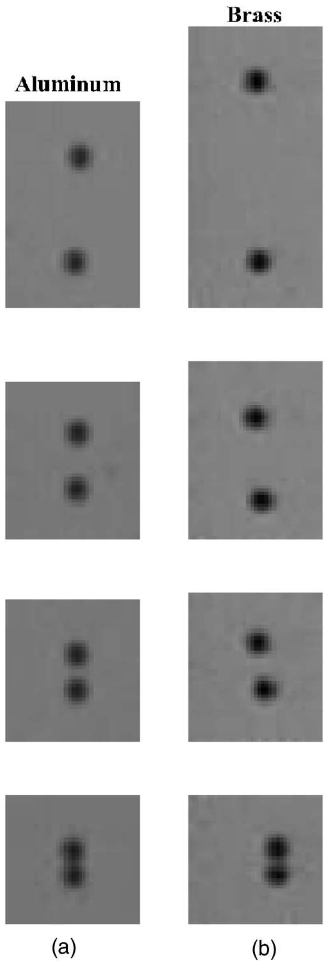

FIG. 2. Snapshots of a doublet of (a) aluminum and (b) brass spheres at successive times showing the aggregation of the particles as they settle. The total width of the images is 9 mm. Both spheres of the aluminum doublet remain on a fixed vertical line in the laboratory's frame whereas, in the same frame, the leading sphere of the brass doublet first moves to the right and is further caught up by the trailing particle.

ogy with the Newtonian case and according to the measured values of v_z, one would expect the z component of the velocity of the leading sphere, $V_{z1}=V_p+v_z(z>0)$ to be larger than the one of the trailing sphere, $V_{z2}=V_p+v_z(z<0)$, resulting in a repulsion rather than in an attraction between spheres. The expected effect would indeed remain weak for the lighter spheres, with velocity differences smaller than 3% of the velocity of a single particle within the vertical distances reported in Fig. 4; in the case of the heavier particles however, the velocity difference thus obtained could reach 8% at a separation distance of four radii and remain close to 5% at a separation distance of height radii. Experimentally, we not only observe opposite behaviors (i.e., faster trailing spheres) but also larger velocity contrasts; the trajectories of Fig. 3 being described within time scales of 100 s for the lighter spheres and 10 s for the heavier ones, the mean relative differences in the velocities are of the order of 10% for the aluminum particles and of 20% for the brass ones.

The attraction between spheres that is observed experimentally is therefore a consequence of the modification of the fluid's state resulting from the passage of the leading sphere: once solicited, the fluid's response to a further solicitation cannot be inferred from the first one. This effect may be attributed to the "memory" of the fluid, as it has been invoked in shear-thinning polymeric fluids [3], but also to the fact that the fluid ages differently according to the stress it is submitted to, as it has been observed in rheological measurements on Laponite suspensions [9]. The observed interaction

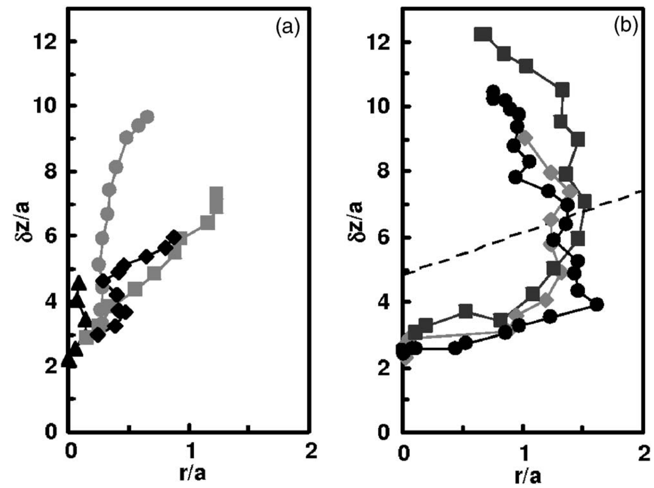

FIG. 3. Trajectories of the trailing sphere of (a) the lighter doublet and (b) the heavier one in the frame of the leading sphere whose center coincides with the origin. The different symbols correspond to experiments with different initial vertical and radial separation between spheres. The spheres eventually form a vertical doublet corresponding to $\delta z=2a$. The dotted line in (b) shows the fluid zero velocity line measured in the single particle's case.

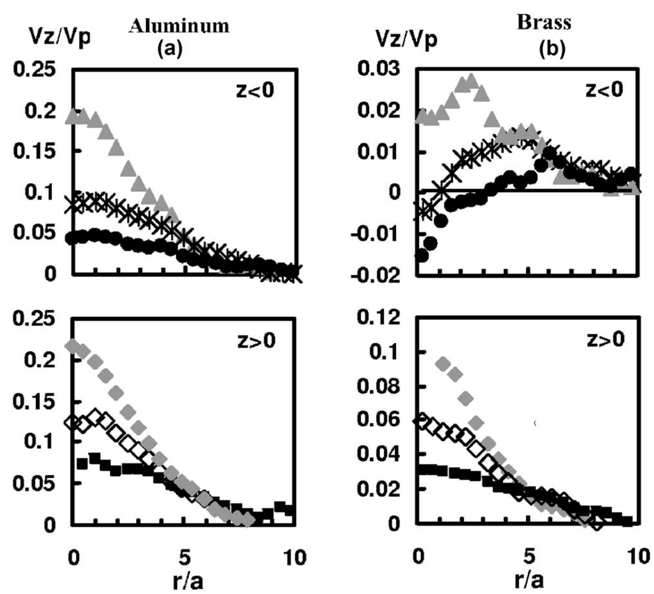

FIG. 4. Vertical component of the fluid velocity as a function of the radial position from (a) an aluminum and (b) a brass sphere. In each case, results upstream ($z>0$) and downstream ($z<0$) are shown at heights from the sphere's center: $z/a=\pm 4$ (full gray symbols), $z/a=\pm 6$ (open symbols) and $z/a=\pm 8$ (full black symbols).

may therefore result from a combination of both properties.

The flow reversal observed in the wake of a single heavier sphere has nevertheless a consequence on the motion of the trailing particle since the relative lateral motion of the particles is observed only in the case of the heavier particles. Furthermore, as shown in Fig. 3, the trailing sphere's trajectory is possibly correlated with the zero velocity line measured in the single sphere's case. In the frame of the leading sphere, the radial velocity of the trailing sphere changes sign at the crossing of this zero velocity line. This unexpected effect had so far never been reported and comparison with experiments performed in other complex fluids is needed in order to further quantify the influence of the negative wake on the two particle behavior.

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