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Oscillations of a solid sphere falling through a wormlike micellar fluid

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We present an experimental study of the motion of a solid sphere falling through a wormlike micellar fluid. While smaller or lighter spheres quickly reach a terminal velocity, larger or heavier spheres are found to oscillate in the direction of their falling motion. The onset of this instability correlates with a critical value of the velocity gradient scale $\Gamma_c \sim 1\ \mathrm{s}^{-1}$. We relate this condition to the known complex rheology of wormlike micellar fluids, and suggest that the unsteady motion of the sphere is caused by the formation and breaking of flow-induced structures.

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A sphere falling through a viscous Newtonian fluid is a classic problem in fluid dynamics, first solved mathematically by Stokes in 1851 [1]. Stokes provided a formula for the drag force F experienced by a sphere of radius R when moving at constant speed V_0 through a fluid with viscosity μ: $F=6\pi\mu R V_0$. The simplicity of the falling sphere experiment has meant that the viscosity can be measured directly from the terminal velocity V_0, using a modified Stokes drag which takes into account wall effects [2]. The falling sphere experiment has also been used to study the viscoelastic properties of many polymeric (non-Newtonian) fluids [3–6]. In general, a falling sphere in a polymeric fluid always approaches a terminal velocity, though sometimes with an oscillating transient [6–8]. In this paper, we present evidence that a sphere falling in a wormlike micellar solution does not approach a steady terminal velocity; instead it undergoes continual oscillations as it falls, as shown in Fig. 1.

A wormlike micellar fluid is an aqueous solution in which amphiphilic (surfactant) molecules self-assemble in the presence of certain organic salts into long tubelike structures, or worms [9]; these micelles can sometimes be as long as 1 μm [10]. Most wormlike micellar solutions are viscoelastic, and at low shear rates their rheological behavior is very similar to that of polymer solutions. However, unlike polymers, which are held together by strong covalent bonds, the micelles are held together by relatively weak entropic and screened electrostatic forces, and hence are constantly breaking and reforming under equilibrium conditions. This provides a new mechanism for stress relaxation during flow [11].

The nonlinear rheology of these micellar fluids can be very different from standard polymer solutions [11–13]. Several observations of new phenomena have been reported, including shear thickening [14,15], a stress plateau in steady shear rheology [16,17], and flow instabilities such as shear banding [16,18], and a shear-induced transition from isotropic to nematic micellar ordering [19]. Chaotic stress fluctuations are also observed in some micellar fluids, for step shear rates above a certain value (in the plateau region of stress-shear rate curve) [20].

There is increasing experimental evidence relating the onset of some of these rheological phenomena to the formation of mesoscale aggregations, or ‘‘shear-induced structures’’ (SIS) [13,21]. These structures have now been imaged directly in wormlike micellar fluids using electron microscopy [22]. Recent experiments have correlated the formation of SIS with the occurrence of shear thickening [15] and shear banding [23]. Also, visualization has shown that the growth of SIS is followed by their tearing or breaking, after which they grow again [15,24].

Recently, nontransient oscillations in the shape and velocity of a rising bubble have been observed in the wormlike micellar system cetyltrimethylammonium bromide (CTAB)/sodium salicylate (NaSal), for concentrations from 8–11 mM [25]. These oscillations occur when the bubble volume is greater than a certain critical value; small bubbles, which remain spherical or ellipsoidal, do not oscillate. The bubble develops a cusp as it rises through the solution. At the moment of cusp formation, the bubble suddenly ‘‘jumps’’ releasing the cusp. A strong negative wake [26] is observed behind the bubble after every jump. Similar oscillatory dynamics are seen in cetylpyridinium chloride/NaSal solutions

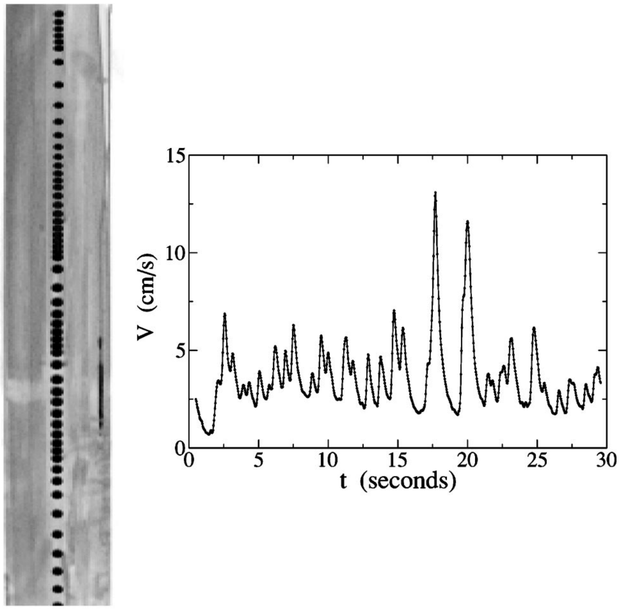

FIG. 1. (Left) Collage of video images showing the descent of a 3/16-inch-diameter teflon sphere in an aqueous solution of 6.0 mM CTAB/NaSal (image shown is 50 cm in height, with $\Delta t = 0.13$ s). (Right) Velocity vs time for a 1/4-inch-diameter Teflon sphere falling through 9.0 mM CTAB/NaSal.

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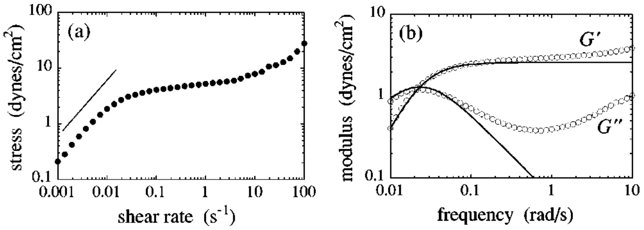

FIG. 2. Shear rheology of 9 mM CTAB/NaSal solution at 30 °C: (a) steady stress vs shear rate. The straight line indicates the linear (Newtonian) scaling; (b) dynamic moduli G' and G'' as functions of frequency. The solid lines are fits to the Maxwell model (see text).

[27], though it is not known if all wormlike micellar fluids will exhibit such a behavior.

The fact that shape oscillations of the bubble are coupled with velocity oscillations suggests that surface tension effects might be responsible. Our observations, that a solid sphere also oscillates while falling through a CTAB/NaSal solution, indicate that such oscillations are not due to surface tension or cusplike tails [25]. We conjecture that the oscillations are due to the formation and destruction of flow-induced structures in the combined shear and extensional flow around the sphere.

Experimental setup and results. Our study focuses on the micellar system CTAB/NaSal [15,16,28], one of several aqueous solutions containing the organic salt sodium salicylate (NaSal), which facilitates the formation of long tubular ‘‘wormlike’’ micelles of cationic surfactants [29]. The CTAB and NaSal used here are obtained from Aldrich, and dissolved in distilled deionized water without further purification. The fluids are mixed for several days, then allowed to settle for a day before use. We restrict our solutions to the molar ratio 1:1 [28]; all data presented here are for 9.0 mM equimolar CTAB/NaSal solutions [except for Fig. 1 (left), which is for 6.0 mM]. At this concentration, the solution is known to form wormlike micelles [15].

The non-Newtonian shear rheology of our fluid was measured using a Rheometrics RFS III concentric Couette rheometer (cylinder inner diameter = 32 mm, height 33 mm, gap 1 mm) at 30 °C. After 100 s of preshear, the steady stress is measured as a function of applied shear rate as shown in Fig. 2(a), with a linear (Newtonian) slope superimposed for comparison. We measure a zero-shear viscosity of 220 P at low shear rates, above which the fluid shear thins. Additionally, a near plateau in stress is observed for shear rates from 0.05 to 2 s^{-1}, a well-known characteristic of wormlike micellar fluids [10,17,29]. Note, however, that the rheology of wormlike micellar fluids can be problematic due to apparent inhomogeneities in Couette [15] and cone and plate rheometers [24,30,31]. The linear viscoelastic rheology is given by the dynamic storage (G') and loss (G'') moduli [5], shown as functions of frequency in Fig. 2(b) for the same apparatus (15% strain). Also shown are fits to a single relaxation time Maxwell model [10,11,19], from which we obtain an elastic modulus $G_0 \simeq 0.26$ Pa, and a relaxation time $\lambda \simeq 43$ s (also given by the crossing frequency $G' \simeq G''$). The increase in G'' at higher frequencies, which deviates from the Maxwell model, is also seen in other wormlike micellar fluids [10].

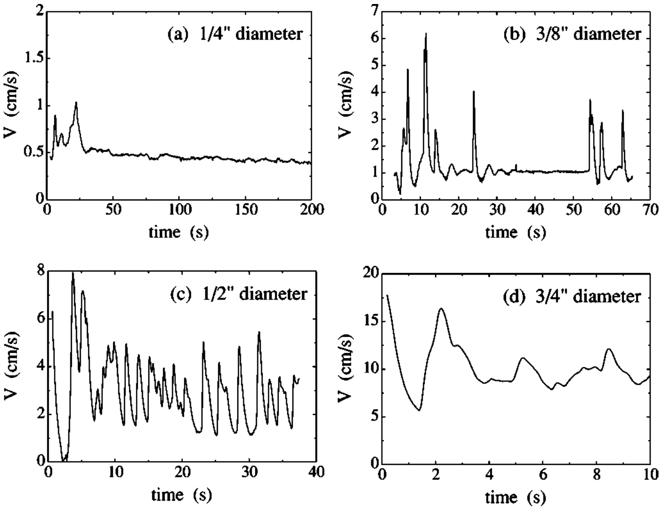

FIG. 3. The velocity of Delrin spheres with diameters of (a) 1/4 inch, (b) 3/8 inch, (c) 1/2 inch, and (d) 3/4 inch.

Our experimental setup consists of a tall cylindrical cell (inner diameter $D = 9$ cm, length $L = 120$ cm) filled with the experimental fluid. For all of our spheres, $d/D \leqslant 0.21$, where d is the sphere diameter. The cylindrical cell is enclosed in a 120-cm-high box, filled with recirculating water to fix the temperature at 30.0 °C. The spheres (made of nylon, Delrin, or Teflon) are dropped in the center of the tube using tweezers, and allowed to fall through 10 cm before data is taken. The fluid was allowed to relax for at least 60 min between each drop. Recovery times much longer than the fluid relaxation time are known to be required even for polymer fluids [3], though here this time may also depend on the decay of micellar structures; we have not studied this effect. The flow around the falling sphere is predominantly viscous and elastic, characterized by low Reynolds numbers ($\mathrm{Re} = Vd/\nu_e \sim 2\times10^{-2}$–10, where V is a typical velocity and ν_e is the effective kinematic viscosity based on the steady rheology), and high Deborah numbers ($\mathrm{De} = \lambda V/d \sim 20$–180); the Stokes number $\mathrm{St} = \rho V d/18\nu_e \sim 2\times10^{-3}$–0.8 (where ρ is the sphere density). The motion of the sphere is captured by a charge-coupled device camera, and stored digitally in a computer. An in-house image analysis program is then used to extract the velocity of the sphere, $V(t)$.

While small spheres reach a terminal velocity after some transient oscillations [Fig. 3(a)], spheres of larger size do not seem to approach a terminal velocity—they oscillate as they fall [Figs. 3(b)–3(d)]. These oscillations are not perfectly periodic, displaying some irregularity. However, an average frequency can be defined as the number of oscillations over the entire fall divided by the time taken. The average frequency of oscillations increases with the radius or volume of the sphere, as was also observed for the oscillations of a rising bubble [25]. Although the amplitude of these velocity oscillations vary widely for a given sphere, the oscillations show a common characteristic of a sudden acceleration and a relatively slower deceleration. At the moment of each acceleration, a strong negative wake [26] is visually observed as a recoil in the fluid, as if the fluid were letting go of the sphere.

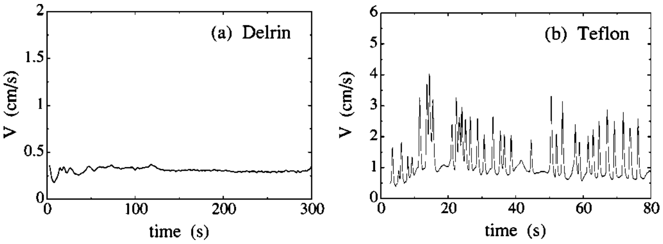

FIG. 4. The velocity of two 3/16-inch-diameter spheres in the same fluid: (a) $\rho = 1.35$ g/cm^3 (Delrin), and (b) $\rho = 2.17$ g/cm^3 (Teflon). The lighter sphere reaches a terminal velocity, whereas the heavier sphere exhibits the unsteady behavior.

For a sphere of fixed radius, there is a transition to oscillations as the density of the sphere is increased. As seen in Fig. 4, a Delrin sphere ($\rho = 1.35$ g/cm^3) with $d = 3/16''$ does not oscillate, while a Teflon sphere ($\rho = 2.17$ g/cm^3) of the same diameter does. However, if the sphere is very large or heavy, it falls faster through the fluid, with less well-defined oscillations [Fig. 3(d)].

To characterize the onset of these oscillations, we estimate the average magnitude of the velocity gradients by the ratio of the velocity of the sphere to its diameter: $\Gamma = V_b/d$, where we take V_b to be the baseline velocity below the oscillations; clearly some measurement of the actual velocity field around the sphere would much improve this estimate. For the four experiments shown in Fig. 3, we find $\Gamma \simeq 0.79$, 1.05, 1.58, and 4.2 s^{-1}, respectively. For Fig. 4, $\Gamma \simeq 0.84$ and 2.1 s^{-1}, respectively. Thus, the oscillations appear to start at a critical value $\Gamma_c \sim 0.9$–1 s^{-1}. This corresponds to the frequency at which G'' begins to increase, and also to a shear rate lying in the nearly flat region of the stress curve (Fig. 2).

Discussion. The first observation of nontransient oscillations of an object in a wormlike micellar fluid was made for a rising bubble [25]. The fact that similar oscillations are observed for a falling sphere indicates that the oscillations are not caused by a surface instability. The sudden acceleration of the sphere during the oscillations indicates that the drag on the sphere has suddenly decreased. We hypothesize that this sudden drop in the drag is due to the break up of flow-induced structures (FIS) that are formed in the region around the sphere.

Using small angle light scattering on a CTAB/NaSal micellar solution in a Couette cell, Liu and Pine found that once the fluid was subjected to shear rates greater than a certain critical value, streaks were observed indicating the presence of mesoscale structures [15]. The formation of such structures, with sizes ~ 1 μm much larger than the diameter of a single micelle (5 nm) [22], dramatically increases both the fluid elasticity and its apparent viscosity [23]. Moreover, the structures do not form instantaneously when the fluid is sheared—they require a finite induction time (order of seconds). These structures grow from the stationary cylinder to the moving cylinder in the Couette cell, but are ripped apart as they approach the moving surface. Then the structures start growing and the cycle begins again. This process can be correlated to macroscopic stress fluctuations, which suggests that FIS cannot sustain large stresses. Although these observations were made for a low concentration shear thickening CTAB/NaSal solution, FIS also form at higher concentrations, such as those seen by Wheeler *et al.* in 40 mM CPCl/NaSal [24].

Based on these observations, we propose a tentative mechanism for the oscillations. First, FIS form around the falling sphere, increasing the effective viscosity and thus the drag. When the stress around the sphere reaches a large enough value, these structures break and the fluid in the wake of the sphere recoils. This causes a sudden drop in the drag experienced by the sphere, which suddenly accelerates. Once the sphere moves into fresh fluid, FIS start forming again, and the cycle repeats.

The critical velocity gradient Γ_c apparently required for the onset of oscillations of a falling sphere suggests a critical gradient required for the formation of FIS. Since the formation of these structures is not a regular, periodic process [15,24], this would explain the irregular oscillations of the sphere. The critical velocity gradient would also explain why smaller or lighter spheres (and small bubbles) do not oscillate; the lower terminal velocity does not shear or stretch the fluid enough to produce the structures. Effectively, lighter spheres move in a more uniform fluid than spheres of moderate weight. On the other hand, very heavy spheres do not have well-defined oscillations [see Fig. 3(d)]. This could be due to the inertia of the sphere, or due to the time needed to form FIS [14,24]; a faster falling sphere may move into fresh fluid before the structures have formed.

Our interpretation also suggests a similar explanation for rising bubbles, where the onset of oscillations coincides with the formation of a cusped tail [25]; the stresses required to form the cusp are also large enough to break the FIS. We predict that every cusped bubble in a micellar fluid which allows for FIS should oscillate. Experiments with bubbles in our CTAB/NaSal solution are so far consistent with this prediction.

Conclusions. A sphere falling in a viscous Newtonian fluid reaches a steady terminal velocity; the approach to this terminal velocity can be shown to be monotonic [32], in agreement with observations. In polymeric fluids, a final steady state is also always observed experimentally. In this paper, we have shown that the unusual behavior seen in the flow of wormlike micellar solutions extends to the classic problem of flow past a sphere. Qualitatively similar oscillations have been reported in another system at large d/D, a dynamically cross-linked guar gum solution [33,34].

We believe that the nontransient oscillations of the falling sphere are caused by the formation FIS. While the rheology of wormlike micellar fluids has typically focused on either pure shear flow or pure extensional flow [35], the velocity field produced by a falling sphere is more complicated; the fluid near the surface is being sheared, whereas in the wake the fluid is primarily under extensional flow. It is thus not a *viscometric flow* [5]. Recently, some evidence of new extensional instabilities has been seen in stretched filaments of wormlike micellar fluids [36]. The observations presented here may be an example of the dynamics of FIS in a complicated hydrodynamic flow.

Analyzing this problem certainly presents a challenge, and mathematical modeling may require a full-scale numerical simulation. Numerical studies of the transient motion of a falling sphere have only recently been undertaken even for polymer fluids (see, e.g., Ref. [7], and references therein). The constitutive equation chosen to model our experiment should give a good fit to the fluid rheology, and, of course, include the as yet unknown mathematical property responsible for the nontransient oscillations.

The shear-stress flow curve for our wormlike micellar fluid displays a flat region [Fig. 2(a)], which is believed to be a manifestation of a nonmonotonic stress-shear rate relation [12,17,18]. It is well known that steady-shear flow in the decreasing region of a nonmonotonic flow curve is unstable (it is ill posed in the Hadamard sense [37]). Such an instability has been attributed to causing different physical effects, such as shear banding in micellar solutions [18], and the shark-skin [37] and spurt [38] instabilities in polymer melts. Heuristically, the oscillations of a falling sphere could be due to the same instability. To test this conjecture, one should choose a constitutive equation which displays a nonmonotonic flow curve, and study the falling sphere problem numerically. Such a simulation is currently in progress.

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