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The Faraday instability in wormlike micelle solutions

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Abstract

The behaviour of semi-dilute wormlike micelle solutions under vertical vibrations is investigated using classical measurements of the Faraday instability threshold along with two new experiments based on particle imaging velocimetry and birefringence. We provide evidence for the presence of oscillations of the critical acceleration and wave number with the vibration frequency, which are linked to oscillations of the velocity and birefringence fields along the vertical direction. These observations are interpreted in terms of standing elastic waves across the fluid layer. Such an interpretation is confirmed numerically by using the model proposed by Kumar [S. Kumar, Vibration-induced interfacial instabilities in viscoelastic fluids, Phys. Rev. E 65 (2002) 026305] for a viscoelastic fluid to calculate the velocity and deformation fields. Finally, further birefringence experiments above the instability threshold show that the Faraday instability does *not* induce disentanglement and breakage of the micellar network.

Keywords: Wormlike micelles; Faraday instability; Particle imaging velocimetry; Birefringence

1. Introduction

In the classical “Faraday experiment” a fluid layer is submitted to vertical vibrations at a frequency f and acceleration a [1]. By increasing the acceleration from zero, the system undergoes a bifurcation from a simple, well-defined state (flat interface with zero velocity in the bulk) to a more complex, destabilized state: at a given acceleration a_c called the critical acceleration, the flat interface goes unstable and a surface wave pattern characterized by the critical wave number k_c develops [1–4]. When the driving acceleration is further increased above a_c, secondary instabilities take place [5], leading at even higher accelerations to spatio-temporal chaos [6]. Together with the Rayleigh–Bénard convective instability, the Faraday instability has often been described as a paradigm for the study of dynamical systems at the laboratory scale [7].

Until about a decade ago, the Faraday instability had been intensively studied in *Newtonian* fluids [4–6,8,9]. Only recently have vertically vibrated complex fluids been the subject of experimental [10–14] and theoretical research [15,16]. It was shown that the coupling between the microstructure of complex fluids and the Faraday instability either only slightly modifies the instability (*e.g.* by affecting the critical acceleration [10,11] or by delaying the transition to disordered states [14]) or presents non-intuitive behaviours such as stabilized “persistent holes” in shear-thickening fluids [12]. With the notable exception of concentrated colloidal and granular suspensions [12,17], most of these works were performed on fluids whose viscoelasticity is only a small perturbation [10,13,14] to Newtonian behaviour. The case of a complex fluid with significant viscoelasticity was addressed in Ref. [11] and it was shown that viscoelasticity could lead to a harmonic response of the fluid surface (*i.e.* at f) instead of the classical subharmonic response (*i.e.* at $f/2$).

In a previous paper [18] we reported onset measurements of the Faraday instability in a semi-dilute surfactant solution (CPyCl–NaSal at 4 wt.%) known to form an entangled network of “wormlike” micelles. In this very viscoelastic fluid, the critical acceleration and wave number were shown to present *oscillations* as a function of the driving frequency f. We interpreted these oscillations in terms of *standing elastic waves* between the disturbed surface and the container bottom.

The present article is devoted to a more thorough study of this striking effect of viscoelasticity on the Faraday instability. After a brief description of our wormlike micellar system and

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of our experimental setup, we first present new sets of Faraday experiments in various wormlike micelle solutions and explore the dependence of the fluid response on surfactant concentration and on temperature. Then, the interpretation in terms of standing elastic waves is confirmed using two novel experiments: particle imaging velocimetry (PIV) and birefringence under vertical vibrations, which show a non monotonic behaviour of the velocity and deformation fields along the vertical direction. We also use the finite-depth model of Ref. [15] to predict the velocity field. This numerical calculation accounts qualitatively for all our experimental observations. Finally, reasons for quantitative discrepancies between the model and the experiments are discussed. In particular, birefringence measurements above onset show that the specific nonlinear features of the rheology of wormlike micelles, namely disentanglement and breakage of the micellar network and shear-induced alignment, do not come into play in our experiments.

2. Viscoelastic fluid: wormlike micelle solution

Wormlike micelles are long (typically hundreds of nanometers to tens of microns), cylindrical (a few nanometers in diameter) and flexible aggregates of surfactant molecules in aqueous solution [19]. Depending on the concentration and on the surfactant, wormlike micelle solutions present various characteristic behaviours: shear-thickenning at low concentration [20], shear-induced alignment in the semi-dilute regime [21], and equilibrium isotropic to nematic phase transition at even larger concentrations [22]. In the following we focus on *semi-dilute* solutions [23]. As shown by Cates [24], the unique viscoelastic behaviour, namely almost a perfect Maxwell fluid [25], of these solutions is due to the interplay of two processes: reptation motion as in conventional polymers [26] and breaking and recombination of the micelles under thermal agitation. Therefore, the complex viscosity of these solutions can be written:

$$\eta^*(\omega) = \eta_0 \frac{1 + \mathrm{i}\omega\tau_2}{1 + \mathrm{i}\omega\tau_1}, \qquad (1)$$

where η_0 is the fluid zero-shear viscosity, τ_1 the relaxation time, ω the pulsation, and τ_2 is a second characteristic time.

For a pure Maxwell fluid, $\tau_2 = 0$. However, due to the presence of the solvent, we shall rather use $\tau_2 = \tau_1\eta_S/\eta_0$, where $\eta_S \ll \eta_0$ is the solvent viscosity (in our experiments, $\eta_S = \eta_{\text{brine}} = 10^{-3}$ Pa s and $\eta_0 \simeq 10$ Pa s). Thus, in the case of a Maxwell fluid, there are really only two free parameters η_0 and τ_1 in Eq. (1) since τ_2 simply derives from the solvent viscosity η_S. Another widely used model for the linear rheology of wormlike micelles is the Oldroyd fluid whose complex viscosity is also given by Eq. (1) but where τ_2 is left as a free parameter (see, *e.g.* refs. [27,28]). We shall see below that this additional free parameter allows one to account for the behaviour of our solutions over a much larger frequency range than the Maxwell fluid. Note that such a use of the Oldroyd model assumes the validity of the Cox–Merz rule which may be violated in some wormlike micelle solutions for $\omega\tau_1 > 1$ [27,29]. We still use fits by the Oldroyd model since Eq. (1) yields good empirical representation of our data. Also note that more elaborate models that account not only for the solvent but also for high frequency relaxation modes based on Rouse or Zimm approaches may apply to wormlike micelle solutions [30]. Finally we introduce the shear modulus as the high-frequency limit of the imaginary part of $-\omega\eta^*(\omega)$, *i.e.* $G_0 = \lim_{\omega\to\infty}\Im(-\omega\eta^*(\omega)) = \eta_0(\tau_1 - \tau_2)/\tau_1^2 \simeq \eta_0/\tau_1$.

In the following, we study various wormlike micelle solutions made of cetylpyridinium chloride (CPyCl, from Aldrich) and sodium salicylate (NaSal, from Acros Organics) dissolved in brine (0.5 M NaCl) with a fixed concentration ratio $[\text{NaSal}]/[\text{CPyCl}] = 0.5$ as described in refs. [21,25]. The CPyCl–NaSal concentration of our solutions varies from 2 to 8 wt.%. This concentration range ensures that (i) the micelles are in the semi-dilute regime, *i.e.* remain entangled and form a viscoelastic network [21] and (ii) the solutions are not too viscous ($\eta_0 \simeq G_0\tau_1 < 100$ Pa s) so that the onset of Faraday waves can be observed.

Linear rheological measurements were performed in the cone-and-plate geometry using a shear rate controlled rheometer (ARES, TA Instruments) with small shear rate oscillations of amplitude 0.1 s^{-1} (the linear regime extends up to at least 1 s^{-1} for all concentrations under study). Such measurements yield reliable values for the viscoelastic moduli $G'(\omega) = -\omega\Im[\eta^*(\omega)]$ and $G''(\omega) = \omega\Re[\eta^*(\omega)]$ as long as the oscillation frequency $f = \omega/2\pi$ remains smaller than about 40 Hz. Fig. 1 shows the

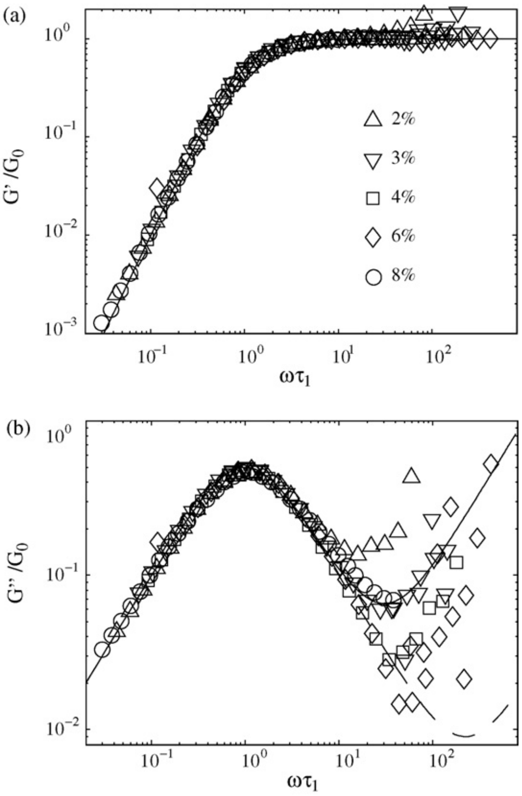

Fig. 1. Linear rheology of 2, 3, 4, 6, and 8 wt.% CPyCl–NaSal solutions. (a) Storage modulus G'/G_0 and (b) loss modulus G''/G_0 vs. $\omega\tau_1$, where ω is the shear oscillation pulsation, τ_1 the main relaxation time, and $G_0 = \lim_{\omega\to\infty} G'(\omega)$ the shear modulus. The dashed line and the continuous line, respectively, represent a Maxwell fluid ($\eta_0 = 49$ Pa s, $\tau_1 = 0.84$ s) and an Oldroyd fluid ($\eta_0 = 50$ Pa s, $\tau_1 = 0.85$ s, $\tau_2 = 0.91$ ms).