

Low-Dimensional Approach to Nonlinear Overstability in the Taylor-Couette Flow of Purely Elastic Fluids

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The finite amplitude purely elastic overstability, for axisymmetric Taylor-vortex flow of highly elastic fluids in the narrow-gap limit, is accurately predicted using the Galerkin projection method. A judicious mode selection is carried out to include the dominant normal stress terms. The resulting twenty-mode dynamical system is capable of capturing the nonlinear behavior observed in the experiment of Muller *et al.* under conditions of negligible inertia. The model predicts, as experiment suggests, the onset of overstability, the growth of oscillation amplitude of flow, and the emergence of higher harmonics in the power spectrum as fluid elasticity increases beyond a critical level. [S0031-9007(97)03432-7]

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Recent linear stability analysis and experiment indicate a dramatic departure in the stability and bifurcation pictures for the Taylor-Couette (TC) flow of viscoelastic fluids, in comparison to Newtonian fluids. While the loss of stability of the circular Couette flow of a Newtonian fluid is inertia driven, that of a viscoelastic fluid can be of purely elastic origin. Viscoelastic fluids tend to exhibit oscillatory Taylor vortex flow (TVF) when the elasticity level exceeds a critical value [1].

The experiment of Muller *et al.* [2] demonstrated, in the case of the TC flow of the so-called Boger fluids [3], the existence of a purely elastic time-periodic instability at a critical rotation rate. The experiment was conducted using laser Doppler velocimetry (LDV), measuring the axial velocity component of a polyisobutylene-based fluid between two concentric cylinders; with the outer cylinder being at rest and the inner cylinder rotating. The results showed an oscillatory flow at a vanishingly small Reynolds number ($Re < 7 \times 10^{-3}$). The flow appeared to undergo a transition from the purely azimuthal Couette flow to time-periodic flow as the Deborah number, De (which is a measure of the relaxation time of the fluid relative to a typical hydrodynamic time scale), exceeded a critical value, De_c . The LDV measurements showed that the oscillatory behavior was not localized, but spread throughout the flow. As De was increased beyond De_c , the amplitude of oscillation increased monotonically. The corresponding power density spectra showed peaks that were instrumentally sharp at the fundamental frequency, the growth of harmonics as De increased, and eventually the emergence of subharmonics as De was further increased. However, more recent experiments by Baumert and Muller [4] seem to suggest that the emergence of subharmonics (or period doubling) may not occur.

The existence of a Hopf bifurcation at $De = De_c$ was established from linear stability analysis of inertialess flow in the narrow-gap [5] and wide-gap [6] limits. The stability of the Hopf bifurcation was later confirmed through the finite-element solution of Northey *et al.* [7]

for the TC flow of an upper-convected Maxwell (UCM) fluid. These authors, however, reported encountering numerical instabilities as De slightly exceeded De_c ; thus, the range of De values for which the solution was obtainable was extremely narrow. More recent numerical calculations were also carried out by Avgousti *et al.* [8] using a pseudospectral method. However, there has been so far no successful direct comparison between theory and experiment on finite-amplitude purely elastic TVF.

It is by now well established that low-order dynamical systems constitute an alternative to conventional numerical methods as one strives to understand the nonlinear behavior of flow [9–14]. Various problems in fluid dynamics have been treated using low-dimensional systems of equations and the theory of nonlinear dynamics [13]. These methods are based on the expansion of the flow field in terms of a complete set of orthogonal functions, Fourier series, or other standard basis functions, and the Galerkin projection technique, which decomposes the initial set of partial differential equations, governing the fluid motion, into an infinite set of ordinary differential equations governing the time-dependent expansion coefficients. The purpose of this Letter is to show that the observed nonlinear dynamics in purely elastic TVF can be effectively described by low-order dynamical systems [9–14]. Attention is focused on the TC flow of the Oldroyd-*B* or Boger type fluid [3] under conditions of negligible inertia, as in the experiment of Muller *et al.* [2]. A polyacrylamide solution in a maltose syrup and water mixture typically constitutes a Boger fluid [15]. The ultimate aim of the model is to recover quantitatively the experimental measurements, and predict what may happen to the flow as De is raised beyond the experimental range.

The interplay between inertia and elasticity for finite-amplitude TVF was previously examined using a similar but severely truncated Galerkin approximation [16]. To make the analysis more tractable, adherence was assumed in the azimuthal direction while slip was assumed along the cylinder axis. In the present model, a judicious mode

selection is carried out in an effort to identify the most influential higher-order normal stress terms (leading to the so-called Weissenberg rod-climbing phenomenon), which were neglected previously, using the more realistic rigid-rigid boundary conditions.

Consider an incompressible viscoelastic fluid of density ρ , relaxation time λ , and viscosity η . The fluid is assumed confined between two infinite and concentric cylinders of inner and outer radii R_i and R_o , respectively. The inner cylinder is taken to be rotating at an angular velocity Ω , while the outer cylinder is at rest. Inertia is neglected. This assumption is usually valid for polymeric flows as viscous effects tend to be dominant. In addition to the mass (continuity) and momentum conservation equations, a suitable constitutive equation must be used.

Although the stability picture is expected to be significantly influenced by the constitutive model adopted [1], the present formulation is restricted to the so-called Boger fluids that obey the Oldroyd-*B* equation [3], similar to the fluid used in the experiment of Muller *et al.* [2]. Some of the properties of Boger fluids are summarized by Larson *et al.* [5]. The test fluid used in the experiment is a dilute solution of a flexible high-molecular-weight polyisobutylene in a viscous low-molecular-weight solvent (polybutene), and is well described by the three-parameter Oldroyd-*B* equation. This constitutive model predicts no shear thinning, a first normal stress coefficient that is constant, and a second normal stress difference that is zero, which is consistent with the rheological properties of dilute solutions [3]. In this case, the excess stress tensor, σ , consists of the sum of a solvent and polymeric solute contributions: $\sigma = \tau - \eta_s \dot{\gamma}$, where $\dot{\gamma} \equiv \nabla \mathbf{u} + (\nabla \mathbf{u})^t$ is the strain-rate tensor, η_s is the solvent viscosity, \mathbf{u} is the velocity, and τ is the elastic part of σ . For a fluid obeying the Oldroyd-*B* equation, one has [17]

$$\lambda \left[\frac{\partial \tau}{\partial t} + \mathbf{u} \cdot \nabla \tau - (\nabla \mathbf{u})^t \cdot \tau - \tau \cdot \nabla \mathbf{u} \right] + \tau = -\eta_p \dot{\gamma}, \quad (1)$$

where η_p is the viscosity of the solute and λ is the relaxation time of the solution. Since inertia is absent, nonlinearities are manifested through the upper-convected terms in Eq. (1).

Consider the flow in the narrow-gap limit, i.e., in the case when the radius R_i/R_o is very close to one. Suitable scales for length, time, velocity, and stress are chosen as for a Newtonian flow [18,19]. One obvious choice is that of the length scale, which, in the present case is taken to be $d \equiv R_o - R_i$. The scaling for $\tau_{\theta\theta}$ needs, however, some careful consideration as it is difficult to estimate its magnitude [1]. In this case, one recovers the important dimensionless groups in the problem, namely, the Deborah number, the solvent-to-polymer viscosity ratio Rv , and the gap-to-radius ratio ε ,

$$\text{De} = \frac{\lambda R_i \Omega}{d}, \quad Rv = \frac{\eta_s}{\eta_p}, \quad \varepsilon = \frac{d}{R_i}. \quad (2)$$

The nonlinear dynamical system is derived by expanding the flow field (velocity, pressure, and stress) into suitably chosen Fourier modes along the axial direction, and symmetric and antisymmetric Chandrasekhar functions [20,21] in the radial direction. The time-dependent expansion coefficients are evaluated by applying the Galerkin projection of the various modes onto the conservation and constitutive equations, and adopting a suitable truncation to close the hierarchy of the resulting twenty-mode nonlinear dynamical system. The most influential normal stress modes are carefully selected to ensure that the relevant dynamics is captured by the approximate model and solution. This is first done by referring to the results from linear stability analysis. The exact solution to the linearized eigenvalue problem is obtained using the direct method, and is compared to the approximate solution based on the Fourier/Chandrasekhar expansion for the eigenvalue problem as in [5]. Since inertia is absent, the linearized equations reduce to a simple constant coefficient equation that is solved analytically. Comparison between the approximate and exact solutions leads to good agreement, especially in the lower wave number range. The more severe truncation level used in our previous work [16] led to a six-dimensional system that was derived by neglecting normal stress terms that tend to become significant for highly elastic flows. The present model takes into account more effectively the influence of normal stresses (which lead to the Weissenberg rod-climbing phenomenon), and is thus adequate for the flow of a highly elastic fluid (with negligible inertia), thus allowing direct comparison with the experiment of Muller *et al.* [2].

Not all experimental flow parameters needed for theory were explicitly reported in Ref. [2]. The test fluid used in the experiment has a (constant) viscosity $\eta = 162$ Pa s, and consists of 1000 ppm of a high molecular weight polyisobutylene, dissolved in a viscous, low molecular weight polybutene of viscosity $\eta_s = 128$ Pa s, so that the solvent-to-polymer viscosity ratio $Rv = 3.76$. The fluid relaxation time λ varies depending on the rheological technique used to measure it. Steady shear flow data give $\lambda = 3.3$ s, while transient relaxation experiments lead to 10.9 s [2,4]. The inner and outer cylinder radii were 8 and 8.5 cm, respectively, so that $\varepsilon = 0.0625$. Although the inner cylinder angular velocity Ω was not explicitly given in the experiment, its value can still be inferred from the values of the experimental Deborah number, De_M , which was introduced by Muller *et al.* [2] as $\text{De}_M = 2\Omega \lambda (1 + \varepsilon)^2 / [(1 + \varepsilon)^2 - 1]$. Note that $\text{De}_{M\varepsilon \rightarrow 0} = \text{De}$. It appears that there was only one fluid used throughout the experiment, and De_M was probably varied by varying only the inner cylinder speed, Ω . Hence, from the range of De_M values reported, the corresponding value of the inner cylinder speed can be given by $\Omega = \text{De}_M / 77.06$ for $\lambda = 4.4$ s. Muller *et al.* [2] reported that the highest Reynolds number, Re , reached in the experiment was of the order 7×10^{-3} . Indeed, if one takes the same definition for the Reynolds number as in Ref. [2], namely,

$Re = \rho \Omega R_i d / \eta$, and considers the value of Ω corresponding to the highest Deborah number reported ($De_M = 54.5$), one finds that $Re = 2.86 \times 10^{-3}$ (assuming the density $\rho \approx 1 \text{ g/cm}^3$). The experimental wave number k (in units of d), at which overstability is first observed, was also not reported by Muller *et al.* [2]; its measurement may have been difficult under transient conditions. Its exact value, however, is not crucial in this case since the critical Deborah number for the onset of overstability does not depend strongly on the wave number, over a wide range of practical values: $k \in [4, 8]$ for $Rv = 3.76$ as linear analysis suggests [5]; this is reflected by the flattening of the corresponding neutral stability curve in the (De, k) plane around the critical value De_c . The wave number will be fixed to $k = 4.85$ for all subsequent calculations. This is the minimum value of the wave number that corresponds to $De_c = 32$ as predicted by the linear stability analysis based on the twenty-mode model. This value is also close to wave numbers reported in other experiments on TC flow of viscoelastic fluids [4]. Thus, as in the experiment of Muller *et al.* [2], only the Deborah number will be varied (by varying Ω) in the following calculations and results.

Consider the flow as De is increased from zero, that is, from the Newtonian level. Linear stability analysis indicates that the Couette flow is unconditionally stable for $De < De_c = 32$. In the absence of inertia, an exchange of stability takes place between the circular Couette flow and oscillatory TVF at the critical Deborah number, since no steady TVF can set in [16]. This is in contrast to a Newtonian fluid in which case only steady TVF sets in at the critical Reynolds number. The main variable of interest is the axial velocity component, $u_z(r, z, t)$, which is obtained from the solution of the twenty-mode nonlinear dynamical system. Here r and z are, respectively, the radial and axial coordinates. In the experiment [2], u_z was measured at the point located midway through the gap, and, probably, where it is maximum over a wavelength in the axial direction. Thus, the amplitude of $u_z(r = (R_i + R_o)/2, z = d/2k, t)$ will be monitored next.

The experimental critical value of the Deborah number, at which oscillatory motion was first detected, was reported to be equal to 32.3, and happens to be slightly larger than the theoretical value $De_c = 32$ predicted by the present linear stability analysis. As De is increased beyond the critical value, calculations show that the amplitude of oscillation increases from zero, confirming the existence and the stability of the Hopf bifurcation, in agreement with experiment [2]. Calculations are carried out for the same range of Deborah numbers as in the experiment: $32 < De < 50$. At $De = 32.5$, the velocity signature and corresponding Fourier spectrum display periodic motion after the purely circular (Couette) flow becomes unstable. The amplitude of oscillation remains relatively small (0.008 cm/s). The power spectrum indicates the presence of a dominant frequency of 0.02 Hz and a weak second harmonics. This periodic behavior persists as De increases, with the flow os-

cillating around the origin (Couette flow). At $De = 43.57$, the motion remains periodic around the origin, with an increase in amplitude to 0.052 cm/s. There is an increase in the fundamental frequency to 0.0298 Hz and the emergence of four significant even and odd harmonics. This trend persists as De is further increased with the eventual emergence of additional harmonics.

Figure 1 shows the Hopf bifurcation for the square of the velocity amplitude based on the twenty-mode model, and the measurements from [2]. Both experiment and theory suggest that the amplitude of oscillation grows like $(De - De_c)^{1/2}$ in agreement with the prediction based on asymptotic analysis in the limit $De \rightarrow De_c$. Figure 2 displays the dependence of the dominant frequency and its harmonics on the Deborah number. The frequency tends to increase with De almost linearly. Unlike the amplitude, the frequency exhibits a jump at the critical Deborah number. This means that any initial weak velocity amplitude at the onset of oscillatory TVF has a dominant frequency that is relatively easy to detect. The agreement between the computed and measured frequencies is obvious from the figure. The apparent growing disagreement for the higher harmonics is to be expected. Any initial discrepancy at the dominant frequency level is simply amplified as it is multiplied by 2 for the second harmonics, by 3 for the third harmonics, and so on.

A closer quantitative agreement between theory and experiment can hardly be envisaged given, on the one hand, the uncertainty surrounding experimental conditions, and, on the other, the lack of a universal and accurate constitutive model for viscoelastic fluids. The sources of discrepancy between theory and experiment are related to limitations for both Newtonian and viscoelastic flow formulations. The lack of a theory capable of predicting the value of the axial wave number k constitutes a major difficulty. The prediction of the value of k remains an

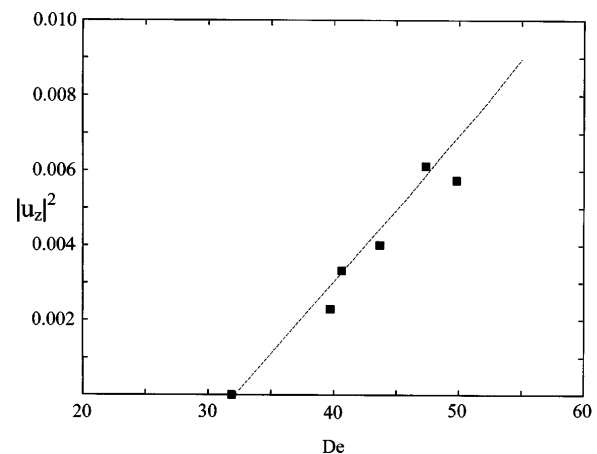


FIG. 1. Bifurcation diagram and comparison between theory (dashed line) and the experiment of Muller *et al.* [2] (squares). The figure shows the square of the axial velocity component amplitude, at $r = (R_i + R_o)/2$ and $z = d/2k$, as a function of the Deborah number.

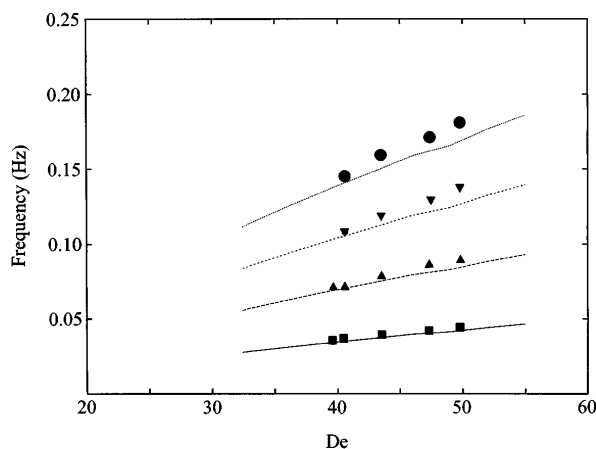


FIG. 2. Frequency of oscillation and comparison between theory (continuous lines) and the experiment of Muller *et al.* (symbols) [2]. The figure shows the dominant frequency and harmonics of the axial velocity component as functions of the Deborah number.

unresolved issue (in a given formulation, it is usually simply imposed from experimental measurement). In the case of viscoelastic Taylor-Couette flow, however, the measurement of k is difficult under transient flow conditions [4]. Other parameters and variables are also difficult to obtain from the experiment of Muller *et al.* [2] and had to be deduced. Additional uncertainty originates from the type of constitutive model used. Although the Oldroyd- B equation predicts the behavior of constant viscosity highly elastic fluids, it does not incorporate the spectrum of relaxation times that is characteristic of real fluids. More complicated constitutive equations, accounting for the nonlinear dependence of the transport coefficients on the rate-of-strain tensor, may also be examined. The present formulation accounts for nonlinearities stemming from the upper-convective terms in the constitutive equation. Another source of discrepancy can come from end effects in the Taylor-Couette apparatus that have been neglected in the present formulation. The narrow-gap approximation is also a limiting assumption. Inertia effects can also play an influential role despite the fact that the experiment was conducted at a vanishingly small Reynolds number ($Re < 10^{-2}$). In general [16], the presence of inertia, no matter how small it may be, prohibits the base flow from losing its stability to the overstable mode. Instead, the base flow loses its stability first to *steady* (and not oscillatory) TVF since there is always a finite range of Re values over which the branches corresponding to steady TVF are stable.

In conclusion, a low-dimensional dynamical system approach is proposed to describe highly elastic TVF. This constitutes a first systematic and accurate theoretical prediction of the purely elastic overstability observed by Muller *et al.* [2]. It is shown that the finite amplitude TVF can be effectively described if higher-order normal stress terms, which were neglected in the previous formu-

lation [16], are properly accounted for. Particularly, the addition of the azimuthal normal stress component leads to additional coupling with higher-order eigenmodes that are of $O(\epsilon De)$. The resulting nonlinear dynamical system involves only twenty degrees of freedom. The sequence of flows predicted by the present model is comparable to that reported by Muller *et al.* [2]. The model predicts the sequence of periodic behaviors observed as the Deborah number is increased: (1) loss of stability of the base flow to an oscillatory flow at a critical Deborah number ($De_c = 32$ as predicted by the model vs 32.3 from experiment), (2) growth of amplitude of the velocity signature like $(De - De_c)^{1/2}$, in agreement with asymptotic analysis, and (3) the emergence of higher harmonics in the Fourier spectrum as De is further increased.

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