Purely Elastic Flow Asymmetries

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(Received 23 April 2007; published 18 October 2007)

Using a numerical technique we demonstrate that the flow of the simplest differential viscoelastic fluid model (i.e., the upper-convected Maxwell model) goes through a bifurcation to a steady asymmetric state when flowing in a perfectly symmetric "cross-slot" geometry. We show that this asymmetry is purely elastic in nature and that the effect of inertia is a stabilizing one. Our results are in qualitative agreement with very recent experimental visualizations of a similar flow in the microfluidic apparatus of Arratia *et al.* [Phys. Rev. Lett. **96**, 144502 (2006)].

DOI: 10.1103/PhysRevLett.99.164503 PACS numbers: 47.50.−d, 05.45.−a, 47.61.−k

Because of their inherent nonlinearity the flow of viscoelastic liquids often leads to strikingly different flow behavior in comparison to the "equivalent" Newtonian flow. Examples of such extreme behavior, often seemingly counterintuitive, include the well-known Weissenberg or rod-climbing effect [1], the Fano or open-siphon effect [2], the large vortex enhancement that is observed in sudden contraction-flow geometries [3], and the extreme off-centerline velocity overshoots that occur in smooth contraction flow [4]. In addition to such gross-flow manifestations of viscoelasticity, a growing body of evidence has shown that viscoelastic fluid flows may develop purely elastic flow instabilities [5]. In contrast, Newtonian liquids flowing at the same conditions remain perfectly steady. Such purely elastic instabilities have been observed experimentally in a number of geometries: Taylor-Couette flow [6], cone-and-plate flow [7], and lid-driven cavity flows [8] amongst many others. It is now well accepted that the destabilizing mechanism which leads to such instabilities is a combination of large normal stresses (which lead to tension along the fluid streamlines) and streamline curvature. McKinley and co-workers [8,9] showed that the curvature of the flow and the tensile stress along the streamlines could be combined to form a dimensionless criterion that must be exceeded for the onset of purely elastic instabilities. More recent experiments have shown that after such purely elastic instabilities set in the flow may go on to develop a chaotic nature that displays many of the same features as classical turbulence [10–12]. In the current Letter we present the first numerical simulations of one such novel viscoelastic instability recently observed experimentally in a "cross-channel" geometry [13].

The birth of "microfluidic" research, and the attendant interest in such flows, has highlighted a number of previously unobserved flow phenomena that occur solely due to viscoelasticity [14]. The inherently small scale of such flows is directly responsible for accentuating the non-Newtonian behavior observed: the small length scale simultaneously makes the Reynolds number ($\equiv \rho U D/\eta$) small and the Deborah ($\equiv \lambda U/D$), or Weissenberg ($\equiv \lambda \dot{\varepsilon}$), number, which characterizes the degree of elasticity in the flow, large. In these expressions ρ and η are the density and shear viscosity of the fluid, respectively, U is a characteristic velocity, D is a characteristic length scale, and $\dot{\varepsilon}$ is the strain rate. Physically the Reynolds number (Re) represents the relative importance of inertial to viscous forces within the flow, the Deborah number (De) represents the ratio of a characteristic time of the fluid (a *relaxation* time, λ) to that of the flow, and the Weissenberg number (Wi) represents the ratio of elastic to viscous stresses in the flow; often for a given flow De and Wi are identical and are used interchangeably. "Elastic turbulence" described above has been exploited to enhance mixing in small curved microfluidic channels [15], while Quake and co-workers [16,17] have exploited the nonlinearity of viscoelastic microfluidic flows to demonstrate devices that can act as memory or control devices such as anisotropic flow rectifiers. An in-depth and systematic study of microfluidic contraction flows [18], in which inertia forces were also present, showed that such flows can also exhibit a wealth of nonlinear dynamics, including inertio-elastic instabilities, large vortex growth, and "diverging" flow [19]. Very recently Arratia *et al.* [13] demonstrated experimentally that the low-Re flow of a flexible polymer solution through a microfluidic cross-channel geometry (which, in earlier macroflow studies was termed a "cross-slot" flow: a convention we adhere to in the current Letter) can give rise to two different instabilities: a first instability in which the flow remains steady but becomes spatially asymmetric and a secondary instability in which the flow becomes unsteady and fluctuates nonperiodically in time. These experimental results were the stimuli for our own numerical investigations that we report here. To our knowledge this first instability, in which the flow remains steady but is strongly asymmetric, is the first time such a steady, possibly purely elastic, asymmetry has been observed experimentally and described in detail. The observations of these various authors, and, in particular, the study of Ref. [13], prompts a natural question: are the instabilities reported amenable to a theoretical explanation [20] or can they at least be predicted by existing constitutive models for viscoelastic fluids?

In the current Letter we show that such a steady asymmetric flow pattern can be predicted using the upper-convected Maxwell model due to Oldroyd [21] in the absence of inertia (i.e., it *is* purely elastic in nature). At low De the flow remains steady and Newtonian-like before, beyond a critical De (≈ 0.31), it becomes bistably asymmetric. With increasing elasticity the flow becomes increasingly asymmetric until beyond a second critical De it eventually becomes time dependent. Although our simulations capture both instabilities, our interest in the current Letter is confined to the first *steady* instability. Our rationale is that this is the first instance that such a steady purely elastic asymmetry has been predicted in contrast to the onset of time dependency, which is more usually observed in creeping flows of viscoelastic fluids at high De.

To model the cross-slot flow, we assume that the flow is creeping (i.e., $\mathrm{Re} \to 0$), two dimensional, isothermal, and incompressible. Under these conditions, the equations that need to be solved are those of conservation of mass $\nabla \cdot \boldsymbol{u} = 0$ and of momentum $-\nabla p + \nabla \cdot \tau = 0$ together with a suitable choice for the viscoelastic stress tensor $\boldsymbol{\tau}$. As already discussed, we use the UCM model [21], $\tau + \lambda \tau_{(1)} = 2\eta \mathbf{D}$, where $\tau_{(1)}$ is the upper-convected derivative and $\mathbf{D}$ is the strain rate tensor. Although a number of shortcomings exist with this model, most notably the unbounded nature of the steady extensional stresses above a critical strain rate and its inability to predict shear-thinning behavior, it is the simplest differential model of an elastic fluid that can capture qualitatively many features of highly elastic flows. A fully implicit finite-volume numerical method is used to solve the constitute equation together with the equations of conservation of mass and momentum. The numerical method has been described in great detail [22,23] and so is not repeated here.

The cross-slot geometry is shown in Fig. 1. The inlet and outlet "arms" are ten channel widths (D) in length, and we impose fully developed velocity (average value U) and stress profiles at inlet and Neumann boundary conditions at the outlets. We confirmed that extending both the outlet arm lengths and mesh refinement had a negligible effect on the critical De and the observed flow patterns.

Streamlines, superimposed upon contours of the first normal-stress difference [scaled using a viscous stress, i.e., $(\tau_{xx} - \tau_{yy})/(\eta U/D)$] to provide an indication of the stress field, are shown for four representative cases in Figs. 2(a)–2(d) for $\mathrm{Re} = 0$. The Newtonian [2(a)] and *low* Deborah number [$\mathrm{De} = 0.3$, 2(b)] results are symmetric, but above a critical Deborah number (≈ 0.31) the flow becomes increasingly asymmetric ($\mathrm{De} = 0.32$ [2(c)] and $\mathrm{De} = 0.4$ [2(d)]). From the numerical results it is possible

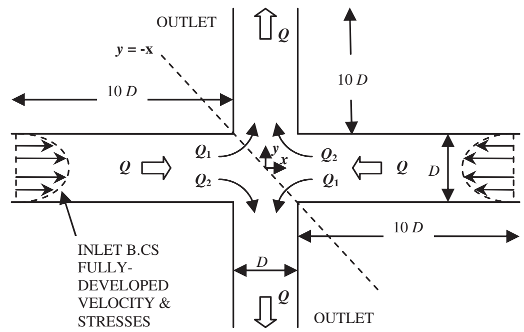

FIG. 1. Schematic of cross-slot geometry. Minimum cell spacing $\Delta x_{\min} = \Delta y_{\min} = 0.01D$, total number of cells = 50601.

to estimate a local Weissenberg number ($\equiv \lambda \dot{\varepsilon}$) based on the strain rate at the stagnation point, and we include these data in Fig. 2 [and as an inset of Fig. 3(b)]. Notably well before the asymmetry sets in, the Weissenberg number at the stagnation point exceeds the "critical" value of 0.5 and therefore the extensional stresses at this point become unbounded. Although singular, the stress field is integrable (our numerical results predict that the τ_{yy} stresses grow to infinity along the horizontal centerline at a rate between $|x|^{-0.25}$ and $|x|^{-0.38}$), and therefore this issue does not prevent a steady solution being obtained. Similar singular behavior is observed in the flow past a salient corner (e.g., in contraction flows) even for creeping flow of a Newtonian fluid [24]. Calculations we have performed using constitutive equations with bounded extensional viscosity (FENE-CR and PTT models), not shown here, also exhibit

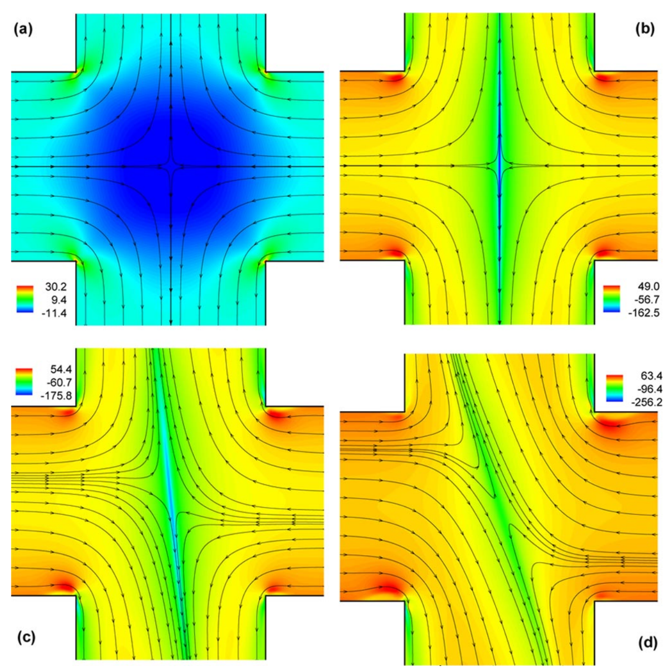

FIG. 2 (color online). Streamline patterns superimposed onto contour plots of $(\tau_{xx} - \tau_{yy})/(\eta U/D)$ to provide an indication of the stress field, for (a) Newtonian fluid, (b) $\mathrm{De} = 0.3$ ($\mathrm{Wi} = 0.57$), (c) $\mathrm{De} = 0.32$ ($\mathrm{Wi} = 0.55$), and (d) $\mathrm{De} = 0.4$ ($\mathrm{Wi} = 0.48$).

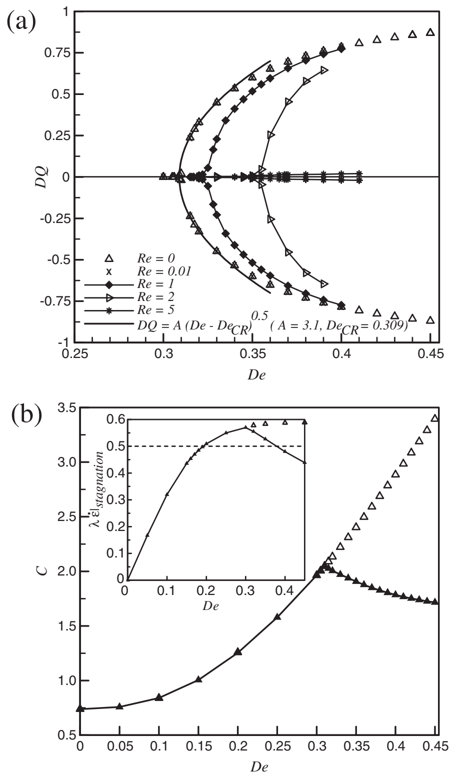

FIG. 3. (a) Growth of asymmetry parameter above critical De for various Re. (b) Variation of Couette correction for full (closed symbols) and symmetry-imposed (open symbols) cases (Re = 0). (b) Inset: Variation of Wi with De (Re = 0).

the asymmetry, and so this singularity is not responsible for the symmetry-breaking bifurcation that we predict. To quantify the degree of asymmetry, we define the parameter $DQ = (Q_2 - Q_1)/(Q_1 + Q_2)$. The total flow rate supplied to each inlet channel $Q = UD = Q_1 + Q_2$ divides into either the upper outflow arm (Q_1 in case of "left" arm) or the lower outflow arm (Q_2 in the case of the left arm) as illustrated in Fig. 1. For a symmetric flow $DQ = 0$ while for a completely asymmetric flow $DQ = \pm 1$. It is also worth emphasizing here that the total flow rate (Q) leaving each outlet arm is the same irrespective of whether the flow is symmetric. We show the variation of this asymmetry parameter with Deborah number in Fig. 3(a). In this figure we also include, close to the critical De, a square root fit to the data which is typical of supercritical bifurcations ($DQ = A\sqrt{\mathrm{De} - \mathrm{De}_{\mathrm{CR}}}$ where $A = 3.1$ and $\mathrm{De}_{\mathrm{CR}} = 0.309$). To understand this phenomenon, we calculate the additional pressure drop that arises due to the presence of the cross slot. To do so we use the so-called "Couette" correction, which is defined as $C \equiv (\Delta p - \Delta p_{\mathrm{fd}})/2\tau_w$ where τ_w is the wall shear stress in the inlet arms evaluated under fully developed conditions and Δp_{fd} is the pressure drop required to drive that fully developed flow in the absence of the slot (i.e., planar channels alone). Thus a unitary Couette correction represents an additional pressure loss equivalent to extending the channel length by one channel width. The variation of C with De is shown in Fig. 3(b) together with the same data for additional calculations in which we have modeled only a quarter of the cross-slot geometry, i.e., imposed symmetry upon the flow. Above the critical Deborah number the additional dimensionless pressure drop is dramatically reduced: thus in bifurcating to a steady asymmetric state the viscoelastic fluid consumes significantly less energy as it flows through the cross slot than it would if it did so symmetrically (at the same De). In bifurcating to the asymmetric state the local Weissenberg number on the stagnation point also decreases [shown as the inset of Fig. 3(b)], and it is likely that this reduction in extensional stresses is linked to the reduction in the pressure drop.

One issue that complicates the comparison between experimental studies and numerical creeping-flow simulations is that, in the former case, inertia will always be present no matter how small. To investigate this finite-inertia effect on the observed asymmetry, we conducted further simulations using the complete momentum equation (i.e., not eliminating the convective term) at Re = 0.01. These results are included in Fig. 3(a), and it is clear that the differences between the truly creeping-flow and the low-Re simulations are negligible. To probe the effects of significant levels of inertia, we performed additional simulations for Re = 1, 2, and 5. As highlighted in Fig. 3(a), these results reveal that the effect of inertia is a stabilizing one in terms of both delaying the onset of the steady bifurcation to higher Deborah numbers and also reducing the magnitude of the asymmetry.

A physical explanation for the observed steady bifurcation can be formulated from the numerical results, and, in particular, by comparing the Newtonian and De = 0.3 solutions with the viscoelastic predictions of higher De (e.g., De = 0.5) under imposed symmetry conditions. In the latter case the flow remains symmetric only because of the imposed boundary conditions (BCs), but the velocity field is already distorted in such a way that if these artificial symmetrical BCs were removed, and the full physical domain used in the simulation, the fluid would respond and evolve to the stable bifurcated state. Inspection of our results reveal that much higher compressive normal stresses are developed by the viscoelastic fluid when the two incoming streams join, which leads to a "turning" velocity profile (into the outlet arm) that is significantly less full, eventually producing a concave, instead of convex, shape, together with an inflection point. Such profile shapes are prone to centrifugal instabilities, as is well known in inner-cylinder-rotating Taylor-Couette flows where Taylor vortices are formed: a fluid element subjected to a perturbation towards a smaller radius of curvature finds itself in a region of faster moving fluid, and the local centripetal pressure gradient tends to push the element even farther towards the center of rotation. Such a mechanism in the case of the cross-slot geometry eventually leads to a steady bifurcated state. Figure 4 shows profiles of the local velocity magnitude along the line $y/x = -1$ (shown as a dotted line in Fig. 1) for three cases: De = 0

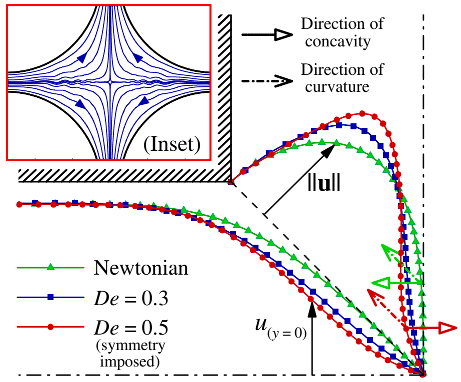

FIG. 4 (color online). Velocity profiles along horizontal centerline and along $y/x = -1$ line (i.e., cross-slot diagonal) for Re = 0 and De = 0, 0.3, and 0.5 (symmetry-imposed) at Re = 0. Inset: A snapshot of the streamlines for a transient simulation for a rounded corner case (De = 2).

(Newtonian case); De = 0.3 (viscoelastic flow immediately prior to bifurcation); and De = 0.5 with imposed symmetry (flow already bifurcated in the complete geometry). The concavity direction of the viscoelastic profile points in the opposite direction to the radius of curvature, thus promoting a centrifugal instability analogous to the velocity distribution in a Taylor-Couette cell with a rotating inner cylinder and the ensuing destabilization mechanism. That the change from convex (stable) to concave (unstable) shape of the velocity profile results from increased compressive stresses is well illustrated by the centerline velocity profile also included in Fig. 4. For the viscoelastic cases the axial velocity starts decreasing earlier and at a faster rate compared to the Newtonian case under the same conditions, on account of the opposing normal forces generated by the frontal collision of the two streams. This feature tends to reduce momentum on the central section of the horizontal channels and, as a consequence of continuity, fluid is pushed towards the outer zones closer to the corners.

In addition to identifying the physical mechanism for the bifurcation to asymmetric flow, our simulations also provide us with a possible triggering mechanism for the appearance of perturbations that eventually lead to the instability. The inset of Fig. 4 shows an instantaneous plot of streamlines when the flow evolves from a stable situation corresponding to a lower De towards an unstable one at higher elasticity albeit in this case in a geometry with rounded corners (specifically it corresponds to a transient flow from rest to De = 2). Sinusoidal oscillations are clearly visible along the horizontal $y = 0$ line close to the joining stagnation point, along which high compressive stresses are generated, while along the vertical line $x = 0$, where the extensional birefringence strand is formed, the vertical streamline remains perfectly straight. Similar simulations without rounded corners tend to mask this effect due to the closer proximity of the sidewalls, which reduce the sharpness of the perturbations and stabilize the flow in the inlet arms. We conclude that perturbations triggering the flow bifurcation result from the joining, compressive flow, along the incoming channels, rather than from the elongational flow emanating vertically along the two outlet channels.

In this Letter we have demonstrated for the first time that a flow asymmetry due solely to elasticity can be numerically predicted in a perfectly symmetric geometry. Our view is that the asymmetry is a consequence of the compressive nature of the flow upstream of the stagnation point rather than the large elongational stresses that also arise in the flow downstream of the stagnation point.

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