Transient drop deformation upon startup of shear in viscoelastic fluids

Pengtao Yue and James J. Feng
Department of Chemical and Biological Engineering and Department of Mathematics,
University of British Columbia, Vancouver, BC V6T 1Z3, Canada

Chun Liu
Department of Mathematics, The Pennsylvania State University, University Park, Pennsylvania 16802

Jie Shen
Department of Mathematics, Purdue University, West Lafayette, Indiana 47907

Recent experiments show that upon abrupt start of a shear flow, a suspended drop undergoes an overshoot in deformation if either the drop or the matrix is a polymeric fluid. Using a diffuse-interface formulation, we carry out two-dimensional numerical simulations that trace the origin of the transient to the mismatch of two time scales: a capillary time for drop deformation and a relaxation time for the polymers in the viscoelastic component. The results are in qualitative agreement with experiments. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2139630]

I. INTRODUCTION

In engineering applications, mixtures and blends are often made of components that are non-Newtonian fluids. Thus, drop dynamics with non-Newtonian rheology in either phase has been studied intensively, both experimentally[1–5] and theoretically.[6–10] Recently, a pair of experimental studies[11,12] focused on the transient reaction of a drop to startup of simple shear when either component is viscoelastic. Sibillo *et al.*[11] measured the transient deformation parameter $D=(L-B)/(L+B)$, L and B being the major and minor axes seen along the vorticity axis, of a Newtonian drop in a Boger fluid matrix. When the capillary number Ca is very small, the deformation D increases gradually and monotonically toward a steady-state value D_s. For larger values of Ca, however, D undergoes an overshoot before settling down to D_s. The drop orientation angle θ initially decreases from 45° as the drop rotates toward the flow direction. Then θ undershoots before reaching the steady orientation. For Ca above the critical value for breakup, D increases monotonically, of course. In all cases, the matrix fluid is a dilute polymer solution that exhibits no stress overshoot when sheared in a rheometer. Thus, the overshoot in D and undershoot in θ for intermediate shear rates must have been due to the interface. The experimental systems of Cherdhirankorn *et al.*[12] are more complex in that both the drop and matrix are molten polymers. The transient deformation D undergoes several cycles of oscillations before stabilizing at the final value.

This paper describes numerical simulations that explore the physical origin of the nonmonotonic drop behavior upon startup of shear flow. We use a diffuse-interface model based on the system's free energy, which accounts for evolving interfaces and non-Newtonian rheology in a unified way.[13,14] By treating the interface as a physically diffuse layer across which properties change continuously, we determine the interfacial position and thickness by a phase-field variable ϕ, whose evolution is governed by Cahn-Hilliard dynamics. This introduces short-range molecular forces and allows to-

pological changes to be simulated naturally as in film rupture.[15] In the current context, however, the phase field is mostly a numerical device for simulating a moving and deforming interface. The polymeric component is modeled as an Oldroyd-B fluid.

II. THEORETICAL AND NUMERICAL MODELS

Yue *et al.*[13] have described the theoretical model and the numerical method in detail, and presented numerical experiments to validate the methodology. Therefore, we only give a brief summary here. For an "immiscible" mixture of a Newtonian fluid and an Oldroyd-B fluid, we introduce a phase-field variable ϕ such that the concentrations of the Oldroyd-B and Newtonian components are $(1+\phi)/2$ and $(1-\phi)/2$, respectively. The interface is simply the level set of $\phi=0$. For simplicity, we take the two components to have the same density though their viscosities may differ. For the governing equations, we first write the usual continuity and inertialess momentum balance: $\nabla \cdot \boldsymbol{v}=0$, $\nabla p=\nabla \cdot \boldsymbol{\tau}$. The total stress tensor is

$$\boldsymbol{\tau}=\left(\frac{1-\phi}{2}\mu_n+\frac{1+\phi}{2}\mu_s\right)[\nabla \boldsymbol{v}+(\nabla \boldsymbol{v})^T]-\Lambda \nabla \phi \nabla \phi$$
$$+\frac{1+\phi}{2}\boldsymbol{\tau}_p, \tag{1}$$

where μ_n is the viscosity of the Newtonian component, μ_s is the viscosity of the Newtonian solvent in the Oldroyd-B fluid, and Λ is the interfacial mixing energy density.[13] The polymer stress $\boldsymbol{\tau}_p$ obeys the Maxwell equation:

$$\boldsymbol{\tau}_p+\lambda_H \boldsymbol{\tau}_{p(1)}=\mu_p[\nabla \boldsymbol{v}+(\nabla \boldsymbol{v})^T], \tag{2}$$

where the subscript (1) denotes the upper convected derivative, λ_H is the polymer relaxation time, and μ_p is the polymer viscosity. The evolution of ϕ, and consequently the interface, is governed by the Cahn-Hilliard equation:

$$\frac{\partial \phi}{\partial t} + \boldsymbol{v} \cdot \nabla \phi = \gamma \Lambda \nabla^2 \left(-\nabla^2 \phi + \frac{\phi(\phi^2 - 1)}{\epsilon^2} \right), \qquad (3)$$

where the mobility parameter γ controls the relaxation time of the interfacial profile and the capillary width ϵ represents the small thickness of the interfacial layer.

We use a spectral method to solve the above equations on a two-dimensional (2D) rectangular domain. Along the flow direction, we impose periodic boundary conditions and use a Fourier expansion. In the transverse direction, we impose Dirichlet conditions with a Chebyshev-Galerkin discretization. The treatment of nonperiodic boundary conditions is numerically nontrivial; it entails splitting the Cahn-Hilliard equation into two Helmholtz equations to which Neumann boundary conditions apply as in Shen.[16] Refinements of grid size and time step have been tested to ensure adequate spatial and temporal resolution, and the numerical code has been validated by comparison with prior results.[13] The Cahn-Hilliard equation, with proper boundary conditions, guarantees mass conservation in the simulation.

FIG. 1. Transient deformation of an Oldroyd-B drop in a Newtonian matrix (O/N) after abrupt start of shear at Ca=0.1. Overshoot in D occurs at sufficiently large De. N/N runs with viscosity ratios λ=1 and 0.5 are also shown for comparison.

III. RESULTS AND ANALYSIS

Initially, a drop of radius a is suspended in the matrix fluid bounded by two solid plates $8a$ apart. The drop is at the center of the channel. At $t=0$, the two plates abruptly start to slide in opposite directions, creating a far-field shear rate of $\dot{\gamma}$. To explore the effect of viscoelasticity on transient drop deformation $D(t)$, we have simulated three cases: a Newtonian drop in a Newtonian matrix (N/N) as the baseline, an Oldroyd-B drop in a Newtonian matrix (O/N), and a Newtonian drop in an Oldroyd-B matrix (N/O). The steady-shear viscosity is matched between the two components except for a few N/N runs with viscosity ratio $\lambda \neq 1$. For the Oldroyd-B component, we fix $\mu_p = \mu_s$. The magnitude of viscoelasticity is represented by the Deborah number De=$\lambda_H \dot{\gamma}$, and the capillary number is defined by Ca=$\mu_m \dot{\gamma}/\sigma$, $\mu_m a$ being the matrix viscosity. Time is scaled by $1/\dot{\gamma}$, length is scaled by a, and the following dimensionless model parameters are used for all the simulations: ϵ=0.04, γ=1.6×10^{-3}.

Analyzing the transient drop deformation requires an understanding of how viscoelasticity inside and outside the drop affects the *steady-state* drop deformation. The steady-state problem has recently been studied by Yue *et al.*[14] For O/N, the polymer molecules are highly stretched near the short-axis of the drop (or the "equator"), and the large tensile stress there engenders a high pressure pushing the equator outward, thereby reducing drop deformation. For N/O, the steady-state D decreases with De first, reaches a minimum, and then increases with De for larger De. The decrease is due to flow modification that reduces the viscous normal stress in the matrix, while the increase is caused by the hoop stress near the equator of the drop where the polymer molecules are stretched the most. These insights will be used in the following to explain the transient drop deformation in terms of the viscoelastic stress field.
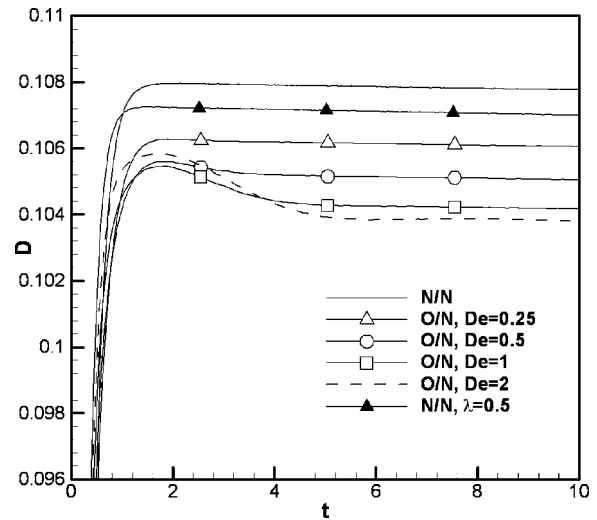
A. A viscoelastic drop in a Newtonian matrix

Figure 1 shows the transient drop deformation $D(t)$ for an Oldroyd-B drop in a Newtonian matrix (O/N) at several De values. Two runs with a Newtonian drop in a Newtonian matrix at different viscosity ratios are also shown for comparison. For large enough Deborah numbers (De>0.25 for these runs), the D curve shows an overshoot, and the amplitude of the overshoot increases with increasing De. It has been suggested[17,3] that such an overshoot may arise from the mismatch of two time scales: the polymer relaxation time λ_H and the capillary time $\tau_\sigma = \mu_m a/\sigma$ for relaxation of the drop shape. This idea will be validated here by tracking the temporal variation of the polymer stress.

The polymer stress inside the drop is known to suppress drop deformation,[14] but it takes a finite time to build up. For two Deborah numbers De=0.25 and 1, Fig. 2 plots the growth of the polymer stress at two points on the drop: the *normal* stress at the tip of the drop and the *tangential* stress at its equator. For the smaller De=0.25, the stress adjusts quickly to the relatively slow drop deformation; at both the tip and the equator the stress grows more quickly than the drop deforms. Thus D increases monotonically in Fig. 1, achieving a D_s below that for N/N. For the larger De=1, the normal stress at the tip develops relatively quickly. But the polymer stretching at the equator is too slow to keep up with the drop deformation. This tensile stress plays the most important role in suppressing drop deformation.[14] The polymer remains mostly coiled during the first phase of drop deformation, contributing little resistance to drop deformation. Only after the drop has deformed significantly does the tensile stress at the equator become large enough to suppress D. Hence the overshoot. D approaches steady state at roughly the same time ($t \approx 6$) as the polymer stress at the equator does. For still higher De, the maximum D increases with De (cf. curve for De=2), evidently because the Oldroyd-B fluid is less viscous initially and offers less resistance to deforma-
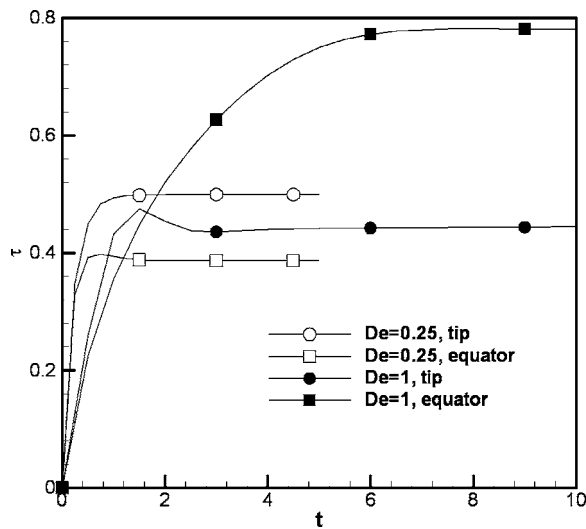
FIG. 2. Growth of the polymer normal stress $\boldsymbol{n} \cdot \boldsymbol{\tau}_p \cdot \boldsymbol{n}$ at the tip and the tangential stress $\boldsymbol{n} \cdot \boldsymbol{\tau}_p \cdot \boldsymbol{t}$ at the equator of the drop for O/N, $\boldsymbol{n}$ and $\boldsymbol{t}$ being the local normal and tangent vectors. Ca=0.1, De=0.25 and 1. The normal stress at the tip and the tangential stress at the equator are the major viscoelastic agents for suppressing drop deformation (Ref. 14). The apparent nonsmoothness of the curves is due to our sampling the solution at large time intervals.

FIG. 3. The deformation of a Newtonian drop in an Oldroyd-B matrix (N/O) at Ca=0.2. $D(t)$ overshoots only for intermediate values of De.

tion. In fact, as De$\rightarrow \infty$, the polymer will contribute little to the stress at any finite time, and we expect the $D(t)$ curve for O/N to approach the N/N curve for a viscosity ratio $\lambda=0.5$, with zero polymer contribution to the viscosity of the Oldroyd-B component.

Note that the time scales for polymer relaxation and drop deformation are represented by De and Ca, respectively. Based on the above analysis, one may naively expect De/Ca>1 to be a criterion for the onset of overshoot in D. The real picture is complicated by several other factors. The drop deformation depends on the viscosity ratio as well; it tends to occur more quickly for smaller λ in Fig. 1, but in any event takes much longer than τ_σ (or Ca in dimensionless terms). Thus, the onset of overshoot appears to be near De/Ca$=5$. More importantly, the polymer stretching, say at the equator, depends on the deformation history of the fluid particles passing through that region, and is not completely determined by the *nominal* De that is based on the far-field shear rate. This point will be revisited toward the end of the paper.

B. A Newtonian drop in a viscoelastic matrix

For a Newtonian drop in an Oldroyd-B matrix, the transient deformation $D(t)$ exhibits an overshoot only for moderate values of De. For the conditions in Fig. 3, one may roughly set this De range as $0.25 < \text{De} < 1$. This behavior can be explained by the same idea of mismatched time scales as for the O/N case. If De is very small, the polymer relaxes very fast and remains more or less in equilibrium with the instantaneous deformation field as the drop deforms and the flow field evolves. There is no overshoot in this case for the same reason that there is no overshoot for small De in O/N. The effect of viscoelasticity here is merely to diminish the final D_s somewhat. If De is large enough so that the polymer
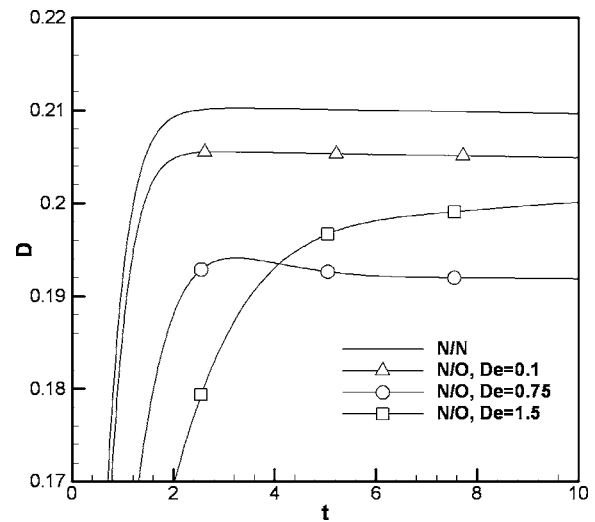
relaxation lags behind the drop retraction, the mismatch in the time scales produces the overshoot as explained before for O/N. This is because for the intermediate De range, viscoelasticity in the matrix in fact acts to *suppress* drop deformation.[14]

As De exceeds the upper bound of this intermediate range (e.g., De=1.5 in Fig. 3), the trend is reversed; the polymer stress in the matrix now enhances drop deformation in the steady state. Therefore, the drop deformation grows monotonically. For still larger De, the polymer unravels so slowly in the matrix that it initially contributes little additional viscosity to the fluid. Consequently, $D(t)$ first reaches a "shoulder" at a level close to that for the same Newtonian drop in a *Newtonian* matrix with just the solvent viscosity (Fig. 4), which is half of the total steady-shear viscosity in this case. Subsequently, the polymer stress increases steadily to very high levels, creating a second phase of drop defor-
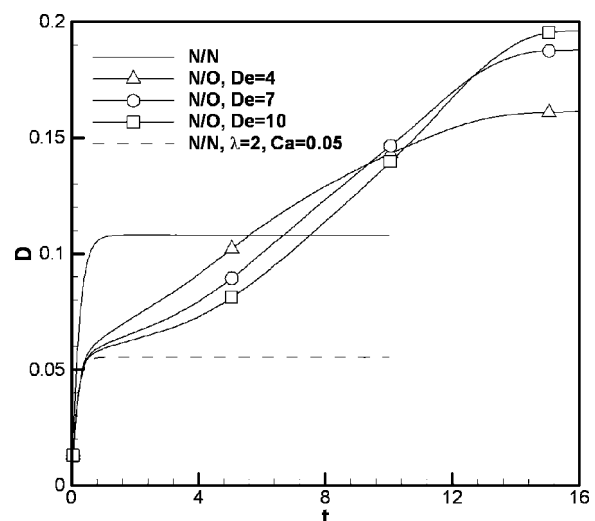
FIG. 4. Two-stage drop deformation for N/O at high De. N/N cases with viscosity ratios $\lambda=1$ and 2 are also shown for comparison. Ca=0.1 for all runs except the N/N case with $\lambda=2$, for which the reduced matrix viscosity yields a smaller Ca.
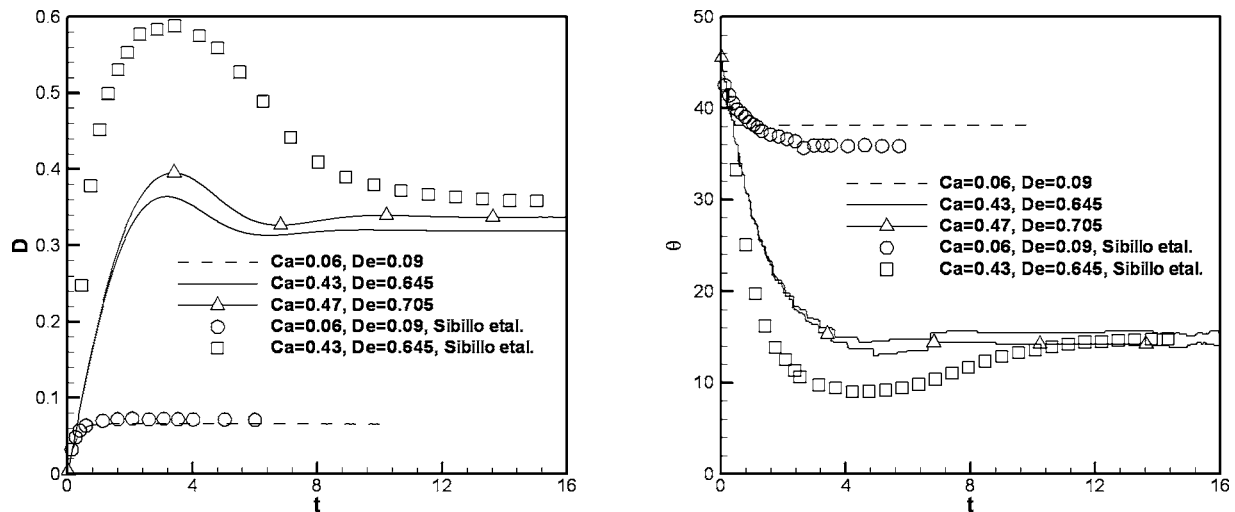
FIG. 5. Comparison between N/O simulations with the experiment of Sibillo *et al.* (Ref. 11). For the experimental data at Ca=0.43, time has been scaled by a factor of 1/4. (a) Transient deformation $D(t)$; (b) transient orientation angle $\theta(t)$.

mation. This phenomenon of two-stage drop deformation has been observed previously by Tretheway and Leal[3] in planar extensional flows. No similar observation has been reported in shear flows.

Numerical experiments indicate that even a Newtonian drop in a Newtonian matrix (N/N) may experience a slight overshoot at relatively high Ca, say Ca$\geqslant$0.5. This corroborates the recent result of Zheng *et al.*[18] However, the overshoot disappears in their 3D simulations, and has apparently never been reported experimentally. In fact, a 3D drop would break at a lower Ca. Within the context of 2D simulations, the N/N overshoot may have originated from competing interfacial modes at high Ca. The viscoelastic overshoot discussed above is due to a distinct mechanism, one that does occur in reality.

We have calculated the transient drop deformation using the experimental parameters of Sibillo *et al.*,[11] and the results are compared in Fig. 5. The experimental runs were carried out by varying the shear-rate for the same fluid pair. Thus, Ca and De vary in proportion. For Ca=0.06 and De=0.09, the simulation and experiment agree almost quantitatively. The drop deforms gradually as in a Newtonian matrix, with no overshoot in the $D(t)$ curve.

For Ca=0.43 and De=0.645, the simulation and experiment both show an overshoot in $D(t)$, but with two important quantitative differences. First, the overshoot takes a much longer time in the experiment than in the simulation. To facilitate visual comparison, we have shrunk t by an arbitrary factor of 4 for this set of experimental data in Fig. 5. Second, the magnitude of the experimental overshoot is much larger than that in our calculation. The different dimensionality might account for part of the differences. Maffettone *et al.*[19] applied a phenomenological ellipsoidal drop model to the transient deformation of a Newtonian drop in a viscoelastic matrix. Their 3D predictions show the same trend as ours: at higher shear rates the overshoot is smaller in magnitude and occurs faster than in experiments. However, the discrepancies do seem to be reduced in 3D. In addition, the fact that there is good agreement for small De suggests another pos-

sibility: the rheology of the experimental fluid may not be adequately represented by the Oldroyd-B model at higher De. Numerical dissipation has been ruled out as a cause for the discrepancy by refining the time steps.

Finally, for Ca=0.47 and De=0.705, the experimental drop has no steady shape; D continues to increase until the drop breaks up (curve not shown). For our 2D simulation, on the other hand, there is no capillary breakup and the $D(t)$ curve merely exhibits a more prominent overshoot. Because their De was not varied independently of Ca, Sibillo *et al.*[11] could not explore higher De values where the two-stage deformation in Fig. 4 might be observed.

Figure 5(b) shows the transient orientation angle $\theta(t)$ of the long axis of the drop. The jitters in the computed curves are a numerical artifact due to interpolation errors in determining the exact location of the drop's tip. Guido *et al.*[4] reported that viscoelasticity in the matrix tends to align the drop more with the flow direction, producing a smaller steady-state θ for larger De. The simulations show the same trend. For Ca=0.06 and De=0.09, the decline of θ is monotonic. For Ca=0.43 and De=0.645, the numerical $\theta(t)$ curve shows a smaller undershoot than the experimental curve. The rotation of the drop toward the flow direction is a consequence of the polymer normal stress near the tip.[4,14] Thus, a more elongated drop will experience a larger torque. This explains the close correspondence between θ and D, including their transients. Incidentally, for O/N the orientational angle hardly changes with drop viscoelasticity, for the obvious reason that there is no viscoelastic torque from the matrix. There is also no overshoot in the orientation angle for O/N.

We have also explored the effect of increasing Ca while keeping De fixed for both N/O and O/N (Fig. 6). This is most conveniently thought of as due to reduction of the interfacial tension with all other parameters unchanged. The magnitude of the overshoot increases with Ca, in apparent contradiction to the idea of competing time scales. A more careful investigation shows that the polymer stress at the equator in fact grows more slowly at higher Ca, and this is due to the
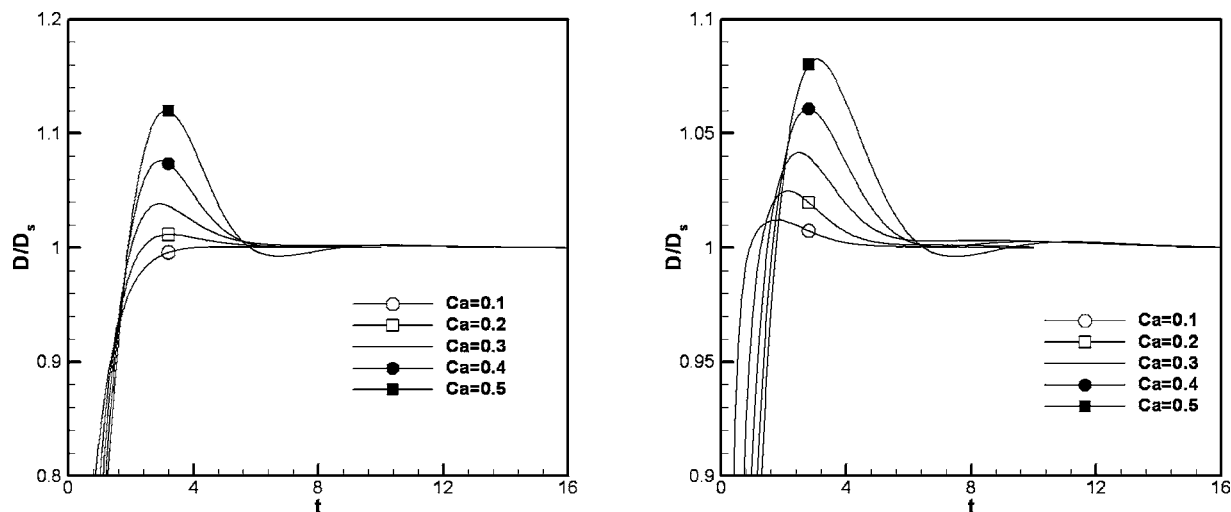
FIG. 6. Increase of the overshoot in D with Ca at a fixed De=0.75. (a) Newtonian drop in Oldroyd-B matrix (N/O); (b) Oldroyd-B drop in Newtonian matrix (O/N).

"weaker" kinematics of the flow, which tends to have more rotation and less elongation as the drop is more elongated. For a small Ca, the drop deviates little from the circular shape. Thus, there is a strong converging flow on the upwind side of the drop.[14] The resulting extension causes the tensile stress near the equator to develop quickly, and may explain the lack of an overshoot for Ca=0.1 in Fig. 6(a). With increasing Ca, the drop becomes more elongated and streamlined, and the extensional component of the flow near the interface is damped down relative to vorticity. Then the polymer stress grows more slowly and the discrepancy in drop and polymer relaxation times is effectively amplified at higher Ca. This bears out the caveat given before on using De/Ca as an indication for the overshoot in D.

IV. SUMMARY

Our diffuse-interface simulations show interesting transients in drop deformation upon startup of shear when either component is viscoelastic. The transients are generally due to the mismatch between the times taken for the drop to deform and for the polymer to stretch. The numerical solutions agree qualitatively with observations,[11] having captured the overshoot in D and undershoot in θ for moderate De. Quantitative discrepancies are attributed to difference in dimensionality and possibly also simplistic representation of the non-Newtonian rheology. For a Newtonian drop in a viscoelastic matrix, the numerical predictions of the disappearance of overshoot for larger De and the two-stage deformation for still larger De are similar to observations in extensional flows,[3] but remain to be confirmed by shear-flow experiments. Although we have restricted viscoelasticity to only one component, it is possible that the same mechanism is at work for mixtures of polymer melts.[12] Simulating such complex systems will certainly require more sophisticated constitutive equations than the Oldroyd-B model used here.

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