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Padé approximants for large-amplitude oscillatory shear flow — Source link

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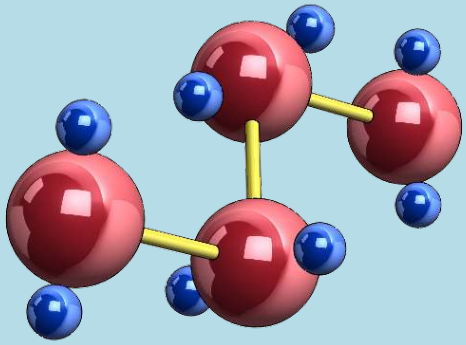

PADÉ APPROXIMANTS FOR LARGE-AMPLITUDE OSCILLATORY SHEAR FLOW

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ABSTRACT

Analytical solutions for either the shear stress or the normal stress differences in large-amplitude oscillatory shear flow, both for continuum or molecular models, often take the form of the first few terms of a power series in the shear rate amplitude. Here we explore improving the accuracy of these truncated series by replacing them with ratios of polynomials. Specifically, we examine replacing the truncated series solution for the corotational Maxwell model with its Padé approximants for the shear stress response, and for the normal stress differences. We find these Padé approximants to agree closely with the corresponding exact solution, and that, in this way, we learn that in this way, one can nearly eliminate the inaccuracies of the truncated expansions.

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I. INTRODUCTION

In certain circumstances, functions can be usefully approximated with ratios of polynomials, and we attribute the method for getting this set of polynomial ratios to Padé ([1]; see Chapter 1 of [2]). Elements of this set are not equally useful, and indeed, some or even all elements will sometimes be of little use. We call each ratio the $[x, y]$ *Padé approximant*, where x is the order of the numerator, and y, of the denominator. Curiously, when the starting function is itself a truncated series, one or more of the Padé approximants may represent the full series more accurately than the starting function (and sometimes, much more accurately), and rheologists sometimes exploit this useful property [3].

Since its conception in 1935 [4,5,6] oscillatory shear flow has become by far the most popular laboratory method for exploring the physics of polymeric liquids. We generate oscillatory shear flow by confining the fluid to a simple shear apparatus, and then subject one solid-liquid boundary to a coplanar sinusoidal displacement, and thus, the fluid to the following velocity profile (see the coordinate system defined and the flow field illustrated in Figure **1**):

$$v_x = \dot{\gamma}^0 y \cos\omega t\,;\; v_y = v_z = 0 \tag{1}$$

and hence, to a corresponding cosinusoidal shear rate:

$$\dot{\gamma}_{yx}(t) = \dot{\gamma}^0 \cos\omega t \tag{2}$$

where, in this paper, symbols are defined in Table II and Table III (which follow Tables 2 and 3 of [7]). Using the characteristic relaxation time of the viscoelastic fluid, λ, we can nondimensionalize Eq. (2):

$$\begin{aligned}\lambda\dot{\gamma}_{yx}(t) &= \lambda\dot{\gamma}^0 \cos\lambda\omega(t/\lambda) \\ &\equiv \mathrm{Wi}\cos\mathrm{De}(t/\lambda)\end{aligned} \tag{3}$$

where the Deborah number is given by:

$$\mathrm{De} \equiv \lambda\omega \tag{4}$$

and, the Weissenberg number, by:

$$\mathrm{Wi} \equiv \lambda\dot{\gamma}^0 \tag{5}$$

The dimensionless Eq. (3) suggests that dimensionless solutions to large-amplitude oscillatory shear flow problems shall be written in terms of Wi or De only, and we follow this throughout this work.

Increasing either the Weissenberg number or the Deborah number in Eq. (3) causes the fluid response to depart from Newtonian behavior. We can construct a complex dimensionless number from the ordered pair $(\mathrm{De},\mathrm{Wi})$ thus (see this illustrated in Fig. 1 of [7]):

$$\mathrm{Gn} \equiv \mathrm{De} + i\,\mathrm{Wi} \tag{6}$$

which defines a vector with magnitude:

$$|\mathrm{Gn}| \equiv \sqrt{\mathrm{De}^2 + \mathrm{Wi}^2} \tag{7}$$

and with angle:

$$\phi \equiv \arctan\frac{\mathrm{Wi}}{\mathrm{De}}. \tag{8}$$

The magnitude of Gn reflects how far the fluid behavior departs from Newtonian behavior. We call such departures *non-Newtonian*. We can associate behavior in steady shear flow with $\mathrm{De}=0$, where $\mathrm{Gn}=i\,\mathrm{Wi}$. We further associate *linear viscoelastic* behavior with $\mathrm{Wi}=0$, where $\mathrm{Gn}\equiv\mathrm{De}$. The angle ϕ thus reflects the type of departure from Newtonian behavior. The value $\phi=0$ corresponds to linear viscoelastic behavior, and $\phi=\pi/2$, to steady shear flow.

When higher harmonics are observed in the shear stress response, we call the oscillatory experiment *large-amplitude*. For polymeric liquids, these higher harmonics are commonly observed when:

$$\frac{\mathrm{Wi}}{\mathrm{De}}>1 \tag{9}$$

which is when the shear stress and the normal stress difference responses generally exhibit higher harmonics. This condition also happens to describe when the measurements become especially challenging [8,9,10,11]. Eq. (9) is thus our working definition of large-amplitude oscillatory shear flow [12,13], and with recent advances in rheometry, conducting experiments satisfying Eq. (9) is now commonplace for exploring the physics of polymeric liquids [14]. The material functions in this flow are most commonly defined as coefficients of the Fourier series:

$$\frac{\tau_{yx}\left(\tau,\gamma_0\right)}{\gamma_0}\equiv-\sum_{\substack{n=1\\ \text{odd}}}^{\infty}G_n'\left(\omega,\gamma_0\right)\sin n\tau+G_n''\left(\omega,\gamma_0\right)\cos n\tau \tag{10}$$

where $\tau\equiv\omega t$. We call this set of coefficients, $G_n'\left(\omega,\gamma_0\right)$ and $G_n''\left(\omega,\gamma_0\right)$, the *Fourier moduli*. These Fourier moduli are readily obtained from a measured time series $\tau_{yx}\left(\tau,\gamma_0\right)$ using the discrete Fourier transform [13]. The notation corresponding to Eq. (10), for the first normal stress difference coefficient, is given in Eq. (179) of [7].

The Fourier moduli in (10) are occasionally expanded in odd powers of γ_0 defining a matrix of frequency dependent nonlinear moduli [15,16,17]:

$$\frac{\tau_{yx}\left(\tau\right)}{\gamma_0}=-\sum_{\substack{m=1\\ \text{odd}}}^{\infty}\sum_{\substack{n=1\\ \text{odd}}}^{m}\gamma_0^{m-1}\left[G_{mn}'\left(\omega\right)\sin n\tau+G_{mn}''\left(\omega\right)\cos n\tau\right] \tag{11}$$

where $\tau\equiv\omega t$.

The shear stress response in LAOS has also been expanded in odd powers of $\dot{\gamma}^0$, defining a matrix of frequency dependent nonlinear viscosities [18]:

$$\frac{\tau_{yx}\left(\tau\right)}{\dot{\gamma}_0}=-\sum_{\substack{n=1\\ \text{odd}}}^{\infty}\sum_{\substack{m=1\\ \text{odd}}}^{n}{\dot{\gamma}^0}^{n-1}\left[\eta_{mn}'\left(\omega\right)\cos m\tau+\eta_{mn}''\left(\omega\right)\sin m\tau\right] \tag{12}$$

where $\left(\eta_{mn}',\eta_{mn}''\right)$ are named the *loss and storage viscosities of mn*th *order*, where $\left(\eta_{11}',\eta_{11}''\right)\equiv\left(\eta',\eta''\right)$. The "$m$" in the "$mn$th order" corresponds to the number of the harmonic in the shear stress response. The "n" in the "mnth order" corresponds

to *one plus* the power of the expansion in Eq. (12). The notation corresponding to Eq. (12), for the first normal stress difference coefficient, is given in Eq. (183) of [7]. Finally, the first normal stress difference response in LAOS has been expanded in odd powers of $\dot{\gamma}^0$, defining a matrix of frequency dependent nonlinear coefficients:

$$\Psi_1(\tau) = -\left[\sum_{\substack{p=1 \\ \text{odd}}}^{\infty} \sum_{\substack{m=0 \\ \text{even}}}^{p+1} \dot{\gamma}^{0\,p-1} \left[\Psi'_{1,m,p+2}(\omega)\cos m\tau + \Psi''_{1,m,p+2}(\omega)\sin m\tau\right]\right] \tag{13}$$

Table I classifies solutions to constitutive equations in large-amplitude oscillatory shear flow for the shear stress that take the form of the truncated power series (Eq. (157) of [7]):

$$\mathbb{S}(\tau) = -A + B\,\mathrm{Wi}^2 - C\,\mathrm{Wi}^4 + \cdots \tag{14}$$

or of Eq. (14) further truncated:

$$\mathbb{S}(\tau) = -A + B\,\mathrm{Wi}^2 - \cdots \tag{15}$$

where each coefficient A, B, C, … is a Fourier series involving odd-valued harmonics. Whereas work on continuum theory employs Eq. (14) (which matches the form of Eq. (28) which is Eq. (58) of [7]), for molecular theory, where calculations are more laborious, Eq. (15) is used (see Eq. (82) of [19]). In this paper, we will compare Eq. (28) with its truncation, Eq. (39), which has the form of Eq. (15). The former comparison will allow us to assess the usefulness of Padé approximants to power series expansions of the form of Eq. (15).

Table I also lists solutions to constitutive equations in large-amplitude oscillatory shear flow for the normal stress differences that take the form of the power series (Eq. (184) of [7]):

$$\mathbb{N}_1(\tau) = -\alpha\,\mathrm{Wi} + \beta\,\mathrm{Wi}^3 - \cdots \tag{16}$$

where each coefficient α, β, … is a Fourier series involving even-valued harmonics. Below we will replace, Eqs. (14), (15) and (16) each with ratios of polynomials whose power series match Eqs. (14), (15) and (16). These ratios are called Padé approximants. To be clear, the Padé approximant is an approximation of the full series from which the first few terms were used as the starting function.

In this paper, we thus explore the usefulness of Padé approximants for the alternant part of the shear stress and normal stress difference responses to the large-amplitude oscillatory shear flow of polymeric liquids. Specifically, we seek to calculate the Padé approximants to truncated expansions for the alternant parts of the shear stress and normal stress differences responses for a corotational Maxwell fluid. The corotational Maxwell model is the simplest model of fluid viscoelasticity that is relevant to large-amplitude oscillatory shear flow, involving just two parameters: η_0 and λ. By *relevant* we mean that the model predicts, as it should, higher harmonics in the alternant parts of the shear stress and the normal stress differences. We select the corotational Maxwell model because it is not only the simplest relevant model, but also, it is the only relevant constitutive model to have yielded an exact solution for the shear stress in large-amplitude oscillatory shear flow [66].

II. ANALYSIS: COROTATIONAL MAXWELL MODEL

For a single-relaxation time, the corotational Maxwell model is:

$$\boldsymbol{\tau} + \lambda \frac{\mathscr{D}\boldsymbol{\tau}}{\mathscr{D}t} = -\eta \dot{\boldsymbol{\gamma}} \tag{17}$$

in which:

$$\frac{\mathscr{D}\boldsymbol{\tau}}{\mathscr{D}t} \equiv \frac{D\boldsymbol{\tau}}{Dt} + \frac{1}{2}\left(\boldsymbol{\omega}\boldsymbol{\tau} - \boldsymbol{\tau}\boldsymbol{\omega}\right) \tag{18}$$

defines the Jaumann derivative; here $D\boldsymbol{\tau}/Dt$ is the substantial derivative and:

$$\dot{\boldsymbol{\gamma}} \equiv \nabla\mathbf{v} + \left(\nabla\mathbf{v}\right)^{\dagger} \tag{19}$$

is the rate-of-strain tensor, and:

$$\boldsymbol{\omega} \equiv \nabla\mathbf{v} - \left(\nabla\mathbf{v}\right)^{\dagger} \tag{20}$$

is the vorticity tensor. We call the derivative given by Eq. (18) *corotational* because it measures rates of changes of the extra stress tensor with respect to a coordinate frame that both translates and rotates with the fluid. For an extensive discussion of corotational models and their applications, see Chapters 7 and 8 of [20], and also [21,22,23,24,25]. The corotational model framework has been closely connected with molecular theory [26,27,28]. For the limiting behaviours of the corotational Maxwell model in steady shear flow, or in the limit of linear viscoelasticity, see Eq. (50), or Eqs. (37) and (47) below.

a. Shear Stress

Here we assess the usefulness of Padé approximants for improving the accuracy of the truncated expansion for the shear stress response of the corotational Maxwell model in large-amplitude oscillatory shear flow whose exact solution is given by [66]:

$$\mathbb{S}(\tau) = -\frac{1}{\mathrm{De}} e^{-\tau/\mathrm{De}} \sin\left(\tfrac{\mathrm{Wi}}{\mathrm{De}} \sin\tau\right) I_1 - \frac{1}{\mathrm{De}} e^{-\tau/\mathrm{De}} \cos\left(\tfrac{\mathrm{Wi}}{\mathrm{De}} \sin\tau\right) I_2 \tag{21}$$

where:

$$I_1 = \mathrm{De}\, e^{\tau/\mathrm{De}} \sum_{k=1}^{\infty} \left[\alpha_k^{(1)} \cos 2k\tau + \beta_k^{(1)} \sin 2k\tau \right] \tag{22}$$

and:

$$\begin{aligned} I_2 = {} & \mathrm{De}\, e^{\tau/\mathrm{De}} \alpha_1^{(2)} \cos\tau + \mathrm{De}\, e^{\tau/\mathrm{De}} \beta_1^{(2)} \sin\tau \\ & + \mathrm{De}\, e^{\tau/\mathrm{De}} \sum_{k=2}^{\infty} \left[\alpha_k^{(2)} \cos(2k-1)\tau + \beta_k^{(2)} \sin(2k-1)\tau \right] \end{aligned} \tag{23}$$

and:

$$\alpha_k^{(1)} \equiv \begin{cases} 0 & ;k=0 \\ \dfrac{-8k^2\,\mathrm{De}^2\,J_{2k}}{\mathrm{Wi}\left(4\,\mathrm{De}^2\,k^2+1\right)} & ;k\geq 1 \end{cases} \tag{24}$$

and:

$$\beta_k^{(1)} \equiv \begin{cases} 0 & ;k=0 \\ \dfrac{4k\,\mathrm{De}\,J_{2k}}{\mathrm{Wi}\left(4\,\mathrm{De}^2\,k^2+1\right)} & ;k\geq 1 \end{cases} \tag{25}$$

and where:

$$\alpha_k^{(2)} = \begin{cases} 0 & ;k=0 \\ \dfrac{J_{2k}+J_{2k-2}}{\mathrm{De}^2\left(2k-1\right)^2+1} & ;k\geq 1 \end{cases} \tag{26}$$

and:

$$\beta_k^{(2)} = \begin{cases} 0 & ;k=0 \\ \dfrac{\mathrm{De}\left(2k-1\right)\left(J_{2k}+J_{2k-2}\right)}{\mathrm{De}^2\left(2k-1\right)^2+1} & ;k\geq 1 \end{cases} \tag{27}$$

By comparing with this exact solution, we can assess how much improvement one might expect from the use of Padé approximants for the shear stress in large-amplitude oscillatory shear flow.

For the corotational Maxwell model, the approximate solution for the dimensionless shear stress response is given by (Eq. (58) of [7]):

$$\mathbb{S}(\tau) = -\frac{\cos\tau + \mathrm{De}\sin\tau}{1+\mathrm{De}^2} + \frac{\mathrm{Wi}^2}{4}\left[\begin{array}{l} \dfrac{3\cos\tau + 6\mathrm{De}\sin\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)} \\ + \dfrac{\left(1-11\mathrm{De}^2\right)\cos 3\tau + 6\left(1-\mathrm{De}^2\right)\mathrm{De}\sin 3\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)} \end{array}\right] - \frac{\mathrm{Wi}^4}{8}\left[\begin{array}{l} \dfrac{5\cos\tau + 15\mathrm{De}\sin\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)} \\ + \left[\dfrac{\left(5-130\mathrm{De}^2\right)\cos 3\tau + \left(45-120\mathrm{De}^2\right)\mathrm{De}\sin 3\tau}{2\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)\left(1+16\mathrm{De}^2\right)}\right] \\ + \left[\dfrac{\left(1-85\mathrm{De}^2+274\mathrm{De}^4\right)\cos 5\tau + \left(15-225\mathrm{De}^2+120\mathrm{De}^4\right)\mathrm{De}\sin 5\tau}{2\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)\left(1+16\mathrm{De}^2\right)\left(1+25\mathrm{De}^2\right)}\right] \end{array}\right] + \cdots \tag{28}$$

which has the form of Eq. (14) where, for the corotational Maxwell model:

$$A \equiv \frac{\cos\tau + \mathrm{De}\sin\tau}{1+\mathrm{De}^2} \tag{29}$$

and:

$$B \equiv \frac{1}{4}\left[\begin{array}{l} \dfrac{3\cos\tau + 6\mathrm{De}\sin\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)} \\ + \dfrac{\left(1-11\mathrm{De}^2\right)\cos 3\tau + 6\left(1-\mathrm{De}^2\right)\mathrm{De}\sin 3\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)} \end{array}\right] \tag{30}$$

and:

$$C \equiv \frac{1}{8}\left[\begin{array}{l} \dfrac{5\cos\tau + 15\mathrm{De}\sin\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)} \\ + \left[\dfrac{\left(5-130\mathrm{De}^2\right)\cos 3\tau + \left(45-120\mathrm{De}^2\right)\mathrm{De}\sin 3\tau}{2\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)\left(1+16\mathrm{De}^2\right)}\right] \\ + \left[\dfrac{\left(1-85\mathrm{De}^2+274\mathrm{De}^4\right)\cos 5\tau + \left(15-225\mathrm{De}^2+120\mathrm{De}^4\right)\mathrm{De}\sin 5\tau}{2\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)\left(1+16\mathrm{De}^2\right)\left(1+25\mathrm{De}^2\right)}\right] \end{array}\right] \tag{31}$$

We examine all Padé approximants of Eq. (14) up to order $[4,4]$ and, in this corner of the Padé table (see Chapters 1 and 2 of [2]), we find exactly four Padé

approximants, that we next list in order of degree, first of their numerators, then, denominators. Approximant of order $[0,2]$ of Eq. (14) is given by:

$$\mathbb{S}(\tau)=\frac{-A^2}{A+B\,\mathrm{Wi}^2} \tag{32}$$

of order $[0,4]$, by:

$$\mathbb{S}(\tau)=\frac{-A^3}{A^2+AB\,\mathrm{Wi}^2-(AC-B^2)\,\mathrm{Wi}^4} \tag{33}$$

of order $[2,2]$, by:

$$\mathbb{S}(\tau)=\frac{-AB+(B^2-AC)\,\mathrm{Wi}^2}{B+C\,\mathrm{Wi}^2} \tag{34}$$

and finally, of order $[2,4]$, by:

$$\mathbb{S}(\tau)=\frac{A^2C-AB^2-\left(2ABC-B^3\right)\mathrm{Wi}^2}{AC-B^2-BC\mathrm{Wi}^2-C^2\mathrm{Wi}^4} \tag{35}$$

Eq. (35) is the first main result of this work. We can also report that this $[2,4]$ approximant exhibits the correct limiting behaviors, for both steady shear flow:

$$\lim_{\mathrm{De}\to 0}\mathbb{S}(\tau)=\lim_{\mathrm{De}\to 0}\frac{A^2C-AB^2-\left(2ABC-B^3\right)\mathrm{Wi}^2}{AC-B^2-BC\mathrm{Wi}^2-C^2\mathrm{Wi}^4}=-\frac{1}{1+\mathrm{Wi}^2} \tag{36}$$

which matches Eq. (84) of [7] (see Eq. (50) below) as it should. For linear viscoelasticity:

$$\lim_{\mathrm{Wi}\to 0}\mathbb{S}(\tau)=\lim_{\mathrm{Wi}\to 0}\frac{A^2C-AB^2-\left(2ABC-B^3\right)\mathrm{Wi}^2}{AC-B^2-BC\mathrm{Wi}^2-C^2\mathrm{Wi}^4}=-\frac{\cos\tau+\mathrm{De}\sin\tau}{1+\mathrm{De}^2} \tag{37}$$

which matches Eq (59) of [7] as it should. It goes without saying that all Padé approximants will be singular where their denominators go to zero, and one must, of course, be mindful of this when we use these. The singularities in Eq. (35) arise, for example, whenever the roots of:

$$AC-B^2-BC\mathrm{Wi}^2-C^2\mathrm{Wi}^4=0 \tag{38}$$

are both real and positive.

In Section III below, we will compare the Padé approximants, Eqs. (32) through (35), with the approximate solution from which these were derived, Eq. (28), and then with the exact solution [Eq. (21) with Eqs. (22)-(27)]. For Padé approximants of Eq. (28), we have limited ourselves to the $[4,4]$ corner of the Padé table, however, were our results unsatisfactory, in any way, we can report that, from experience, extending our search area to a larger corner of the Padé table does not help. By *unsatisfactory*, we at least mean, when the shear stress loops lack two-fold symmetry. In our subsequent treatment of Eq. (39), we get less satisfactory results. Given the usefulness of our $[2,4]$ approximant, we see no reason to pursue orders exceeding $[4,4]$.

Mindful of the discussion following Eq. (15) above, we next truncate Eq. (28) to its next lowest power of Wi:

$$\mathbb{S}(\tau) = -\frac{\cos\tau + \mathrm{De}\sin\tau}{1+\mathrm{De}^2} + \frac{\mathrm{Wi}^2}{4}\left[\begin{array}{l} \dfrac{3\cos\tau + 6\mathrm{De}\sin\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)} \\ + \dfrac{\left(1-11\mathrm{De}^2\right)\cos 3\tau + 6\left(1-\mathrm{De}^2\right)\mathrm{De}\sin 3\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)} \end{array}\right] + \cdots \tag{39}$$

which has the general form of Eq. (15). We examine all Padé approximants of Eq. (15), up to order $[4,4]$ and we find exactly two Padé approximants, which we next list in order of degree, first of their numerators, then, denominators. Approximant $[0,2]$ matches Eq. (32), and approximant $[0,4]$ is given by:

$$\mathbb{S}(\tau) = \frac{-A^3}{A^2 + AB\,\mathrm{Wi}^2 + B^2\mathrm{Wi}^4} \tag{40}$$

which is just Eq. (33) evaluated for $C = 0$. We will examine Eqs. (39) and (40) in Section III below.

b. Normal Stress Differences

Here we assess the usefulness of Padé approximants for improving the accuracy of the truncated expansion for the normal stress difference responses of the corotational Maxwell model in large-amplitude oscillatory shear flow. We select the corotational Maxwell model because it is not only the simplest relevant model, but also, it is the only relevant constitutive model to have yielded an exact solution for the normal stress differences in large-amplitude oscillatory shear flow. This exact solution is given by:

$$\mathbb{N}_1(\tau) = -2\mathbb{N}_2(\tau) = \frac{2}{\mathrm{De}} e^{-\tau/\mathrm{De}} \cos\left(\tfrac{\mathrm{Wi}}{\mathrm{De}}\sin\tau\right) I_1 - \frac{2}{\mathrm{De}} e^{-\tau/\mathrm{De}} \sin\left(\tfrac{\mathrm{Wi}}{\mathrm{De}}\sin\tau\right) I_2 \tag{41}$$

where I_1 and I_2 are given by Eqs. (22) and (23). By comparing with this exact solution, we can assess exactly how much improvement one might generally expect from the use of Padé approximants for the normal stress differences in large-amplitude oscillatory shear flow.

For the corotational Maxwell model, the dimensionless normal stress differences are given by (Eq. (66) of [7]):

$$\begin{aligned}\mathbb{N}_1(\tau) &= -2\mathbb{N}_2(\tau) \\ &= -\mathrm{We}\left[\frac{1}{1+\mathrm{De}^2}+\frac{\left(1-2\mathrm{De}^2\right)\cos 2\tau+3\mathrm{De}\sin 2\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)}\right] \\ &+\frac{\mathrm{We}^3}{4}\left[\begin{array}{l}\dfrac{3}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)}+\dfrac{\left(4-24\mathrm{De}^2\right)\cos 2\tau+20\mathrm{De}\sin 2\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)} \\ +\dfrac{\left(1-35\mathrm{De}^2+24\mathrm{De}^4\right)\cos 4\tau+\left(10\mathrm{De}-50\mathrm{De}^3\right)\sin 4\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)\left(1+16\mathrm{De}^2\right)}\end{array}\right]\end{aligned} \tag{42}$$

which has the form of Eq. (16) where:

$$\alpha \equiv \left[\frac{1}{1+\mathrm{De}^2}+\frac{\left(1-2\mathrm{De}^2\right)\cos 2\tau+3\mathrm{De}\sin 2\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)}\right] \tag{43}$$

and:

$$\beta \equiv \frac{1}{4}\left[\begin{array}{l}\dfrac{3}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)}+\dfrac{\left(4-24\mathrm{De}^2\right)\cos 2\tau+20\mathrm{De}\sin 2\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)} \\ +\dfrac{\left(1-35\mathrm{De}^2+24\mathrm{De}^4\right)\cos 4\tau+\left(10\mathrm{De}-50\mathrm{De}^3\right)\sin 4\tau}{\left(1+\mathrm{De}^2\right)\left(1+4\mathrm{De}^2\right)\left(1+9\mathrm{De}^2\right)\left(1+16\mathrm{De}^2\right)}\end{array}\right] \tag{44}$$

We examine all Padé approximants of Eq. (16) up to order $[4,4]$ and we find exactly two Padé approximants, that we next list in order of degree, first of their numerators, then, denominators. Approximant of order $[1,2]$ of Eq. (16) is given by:

$$\mathbb{N}_1(\tau) = -\frac{\alpha^2\,\mathrm{Wi}}{\alpha+\beta\,\mathrm{Wi}^2} \tag{45}$$

and of order $[1,4]$, by:

$$\mathbb{N}_1(\tau) = \frac{-\alpha^3\,\mathrm{Wi}}{\alpha^2+\alpha\beta\,\mathrm{Wi}^2+\beta^2\mathrm{Wi}^4} \tag{46}$$

Eq. (46) is the second main result of this work. We can also report that this $[1,4]$, approximant the correct limiting behavior for linear viscoelasticity:

$$\lim_{\mathrm{Wi}\to 0}\mathbb{N}_1(\tau) = \lim_{\mathrm{Wi}\to 0}\frac{-\alpha^3\,\mathrm{Wi}}{\alpha^2+\alpha\beta\,\mathrm{Wi}^2+\beta^2\,\mathrm{Wi}^4} = 0 \tag{47}$$

as it should, and that for its behavior in steady shear flow, we get:

$$\lim_{\mathrm{De}\to 0}\mathbb{N}_1(\tau) = \lim_{\mathrm{De}\to 0}\frac{-\alpha^3\,\mathrm{Wi}}{\alpha^2+\alpha\beta\,\mathrm{Wi}^2+\beta^2\,\mathrm{Wi}^4} = \frac{-2\,\mathrm{Wi}}{1+\mathrm{Wi}^2+\mathrm{Wi}^4} \tag{48}$$

or that:

$$\frac{\Psi_1}{2\eta_0\lambda} = \frac{1}{1+\mathrm{Wi}^2+\mathrm{Wi}^4} \tag{49}$$

which is only a close approximation to Eq. (84) of [7]:

$$\frac{\eta}{\eta_0} = \frac{\Psi_1}{2\eta_0\lambda} = \frac{1}{1+\mathrm{Wi}^2} \tag{50}$$

and so the limiting behavior of Eq. (46), though qualitatively correct, is only approximately right. Since the denominator in Eq. (46) features no real roots, Eq. (46) presents no problematic singularities.

In Section III below, we will compare the Padé approximants, Eq. (45) and (46), with the approximate solution from which this was derived, Eq. (42), and then with the exact solution [Eq. (41) with Eqs. (22)-(27)]. Exploring the Padé table beyond the order $[4,4]$ may uncover more accurate, though no less complex, approximants. Given the usefulness of our $[1,4]$ approximant, we chose not to investigate orders exceeding $[4,4]$.

III. RESULTS

When using the exact solutions given by either Eq. (21) or Eq. (41) with approximate solutions, one must decide on the number of terms to keep in the Bessel functions, and also on how many harmonics to keep in the stress calculations. For the Bessel functions, we find 40 terms to be more than sufficient for our results to be at least invariant to within a line width for all of the figures reported in this paper, and so we kept 40 terms for the Bessel functions throughout. For our figures, we use a computer to evaluate the exact solutions given by Eqs. (21) or (41). For our alternant loop evaluations, we included 40 harmonics, and coded Eqs. (41) and into MATLAB (Version R2012b). On a MacBook Air (1.3 GHz Intel Core i5 processor with 4GB 1600 MHz DDR3 memory) employing the OS X (Version 10.9.5) operating system, we find such an evaluation to consume less than 5 minutes of CPU time. Keeping 40 terms in the Bessel functions, with 40 harmonics in the Fourier series represents an abundance of caution. In all of our figures below, the loops for the exact solution have thus been calculated to a precision that falls well below the line widths.

Figure 2 through Figure 5 illustrate the improvements in the predictions for the shear stress that we realize when we covert Eq. (28) to its $[0,2]$, $[0,4]$, $[2,2]$ and $[2,4]$ Padé approximants given by Eqs. (32) through (35). In Figure 5, we discover stunning improvement upon Eq. (28) with the $[2,4]$ approximant, which agrees with the exact solution [Eq. (21) with Eqs. (22)-(25) and with (23)-(27)]. By *stunning*, we mean that although Eq. (28) involves only up to the fifth harmonic (and then only a first approximation of this fifth), the $[2,4]$ approximant of Eq. (28) agrees closely with the exact solution, which, of course, contains all of the higher harmonics. We learn that with the use of Padé approximants, for the shear stress in large-amplitude oscillatory shear flow, one can take a power series expansion in the shear rate amplitude, truncated after the fourth power of the Weissenberg number (as thus, truncated after the first term

to contain the fifth harmonic), and then get loops that agree closely with the exact solution (even under extreme conditions for the Weissenberg number). Since obtaining the first few terms of a power series in the shear rate amplitude is laborious, even for the simplest relevant constitutive models [7,19,29,30], we recommend the use of Padé approximants to get the most out of this labor.

From Table I we learn that solutions of the form of Eq. (15) are much more commonly available that of Eq. (14). In this sense, the successful Padé approximation of Eq. (15) would be of wider use. Figure 6 and Figure 7 illustrate the improvements in the predictions for the shear stress that we realize when we convert Eq. (39) [which has the form of Eq. (15)] to its $[0,2]$ and $[0,4]$ Padé approximants given by Eqs. (32) and (40). In Figure 7, we find satisfactory agreement with the exact solution for the shear stress [Eq. (21) with Eqs. (22)-(27)], but the result is not stunning. Specifically, the $[0,4]$ approximant yields a little two-fold asymmetry in the shear stress loops, which makes the prediction slightly unphysical. Otherwise, the agreement with the exact solution is close.

Figure 8 and Figure 9 illustrate the improvements in the predictions for the normal stress differences that we realize when we convert Eq. (42) [which has the form of Eq. (16)] to its $[1,2]$ and $[1,4]$ Padé approximants given by Eqs. (45) and (46). In Figure 9 we find satisfactory agreement with the exact solution for the normal stress differences [Eq. (41) with Eqs. (22)-(27)], but the result is not stunning. Specifically, the $[1,4]$ approximant yields a huge improvement over Eq. (42), shares the same shape as the exact solution, but falls just below the exact solution.

From Figure 7 or Figure 9 of our results, we learn that even for the most highly truncated forms published for analytical solutions, which are always arrived laboriously, it pays to use Padé approximants when using theory or when comparing theory with experimental measurements. Here, specifically, we have examined Eqs. (39) and (42) for the corotational Maxwell model as examples of the truncated forms, Eqs. (15) and (16).

IV. CONCLUSION

This work begins by noticing, from our examination of Table I, that analytical solutions to large-amplitude oscillatory shear flow problems often take the form of truncated expansions in the shear rate [Eqs. (14), (15) and (16)]. Recognizing that getting even the first few terms of such expansion, for even the simplest of relevant constitutive models [7,19,29,30], is laborious. We were thus motivated to devise a method to help make the most of the three common truncations of these expansions (for both the shear stress and the normal stress differences) using Padé approximants.

For the shear stress, when the expansion is truncated after the fourth power of the Weissenberg number, we find stunning accuracy in the $[2,4]$ Padé approximant (see Figure 5). When the expansion is truncated after the second power of the shear rate amplitude (as is more commonly done), we find satisfactory accuracy in the $[0,4]$ Padé approximant (see Figure 7). For the normal stress differences, when the expansion is truncated after the third power

of the Weissenberg number, we find satisfactory accuracy in the $[1,4]$ Padé approximant (see Figure 9).

From this work, we learn that even for the most severely truncated forms published for analytic solutions, it pays to use Padé approximants when evaluating theory or when comparing it with experimental measurements.

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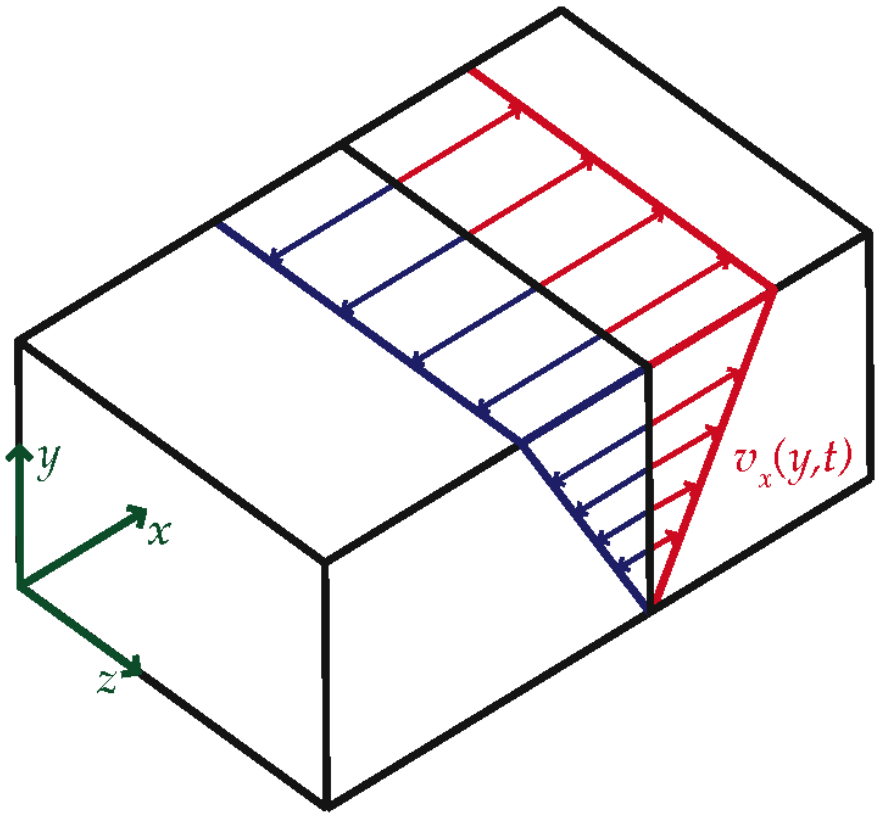

Figure 1: Orthomorphic isometric sketch of alternating velocity profile in oscillatory shear flow [Eq. (1)]. Cartesian coordinates with origin on the stationary plate. The linear velocity profile results from the assumption that inertial effects can be neglected.

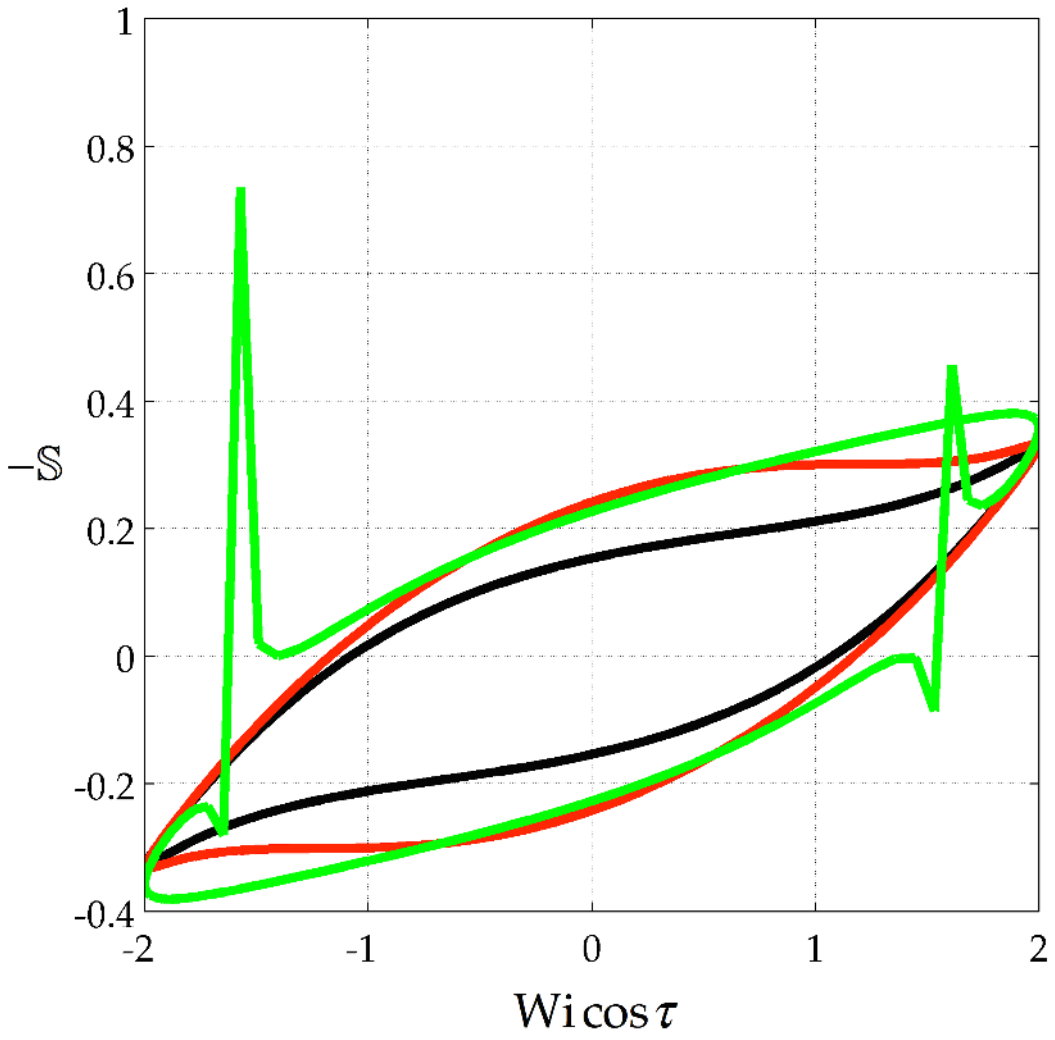

Figure 2. $[0,2]$ Padé approximant of Eq. (28) (**green**) *versus* Eq. (28) (**red**) and also *versus* exact solution [Eq. (21) with Eqs. (22)-(27)] (**black**) for alternance. Counterclockwise loops of minus dimensionless shear stress, $-\mathbb{S}$, *versus* dimensionless shear rate, $\lambda\dot{\gamma}$, calculated for the 2-constant corotational Maxwell model with $\mathrm{Wi}/\mathrm{De} = 2$ and $\lambda\omega = 1$.

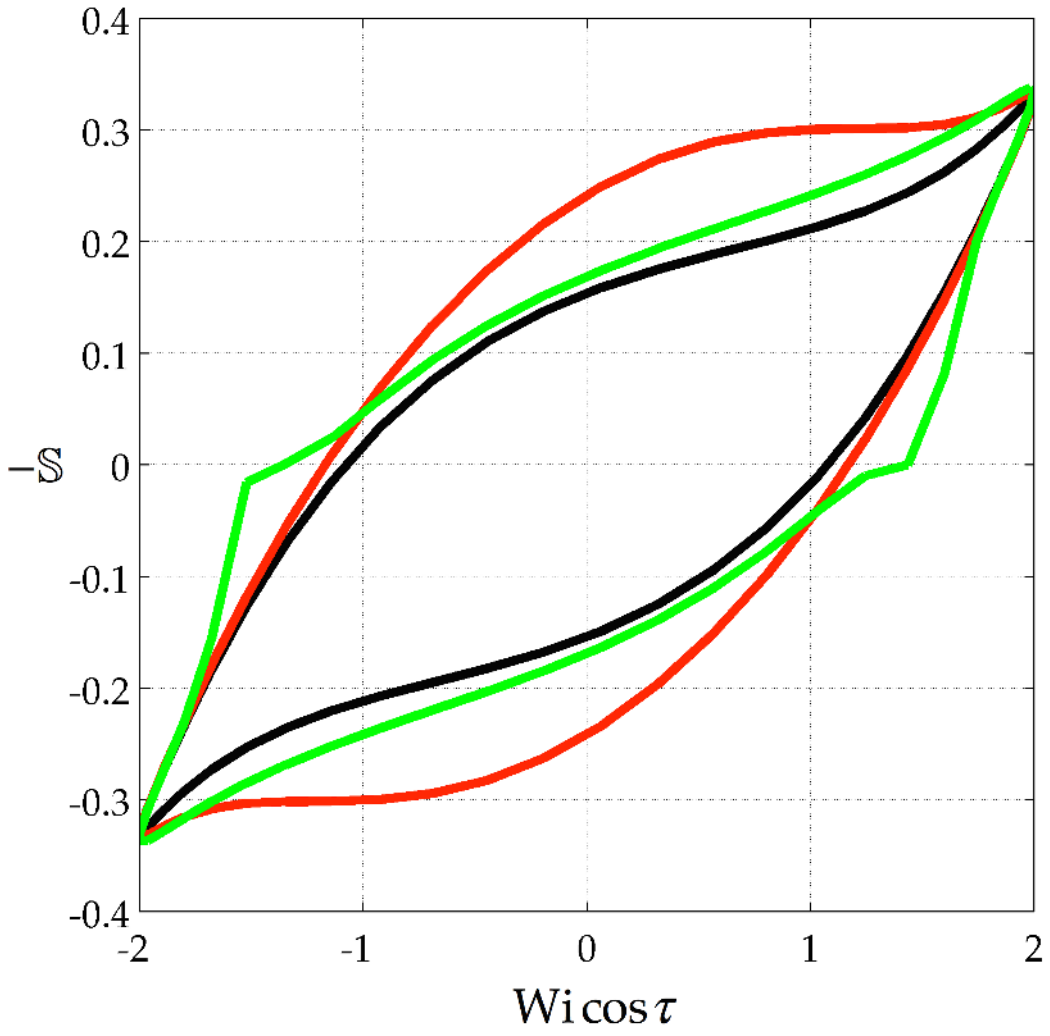

Figure 3. $[0,4]$ Padé approximant of Eq. (28) (**green**) *versus* Eq. (28) (**red**) and also *versus* exact solution [Eq. (21) with Eqs. (22)-(27)] (**black**) for alternance. Counterclockwise loops of minus dimensionless shear stress, $-\mathbb{S}$, *versus* dimensionless shear rate, $\lambda\dot{\gamma}$, calculated for the 2-constant corotational Maxwell model with $\mathrm{Wi}/\mathrm{De} = 2$ and $\lambda\omega = 1$.

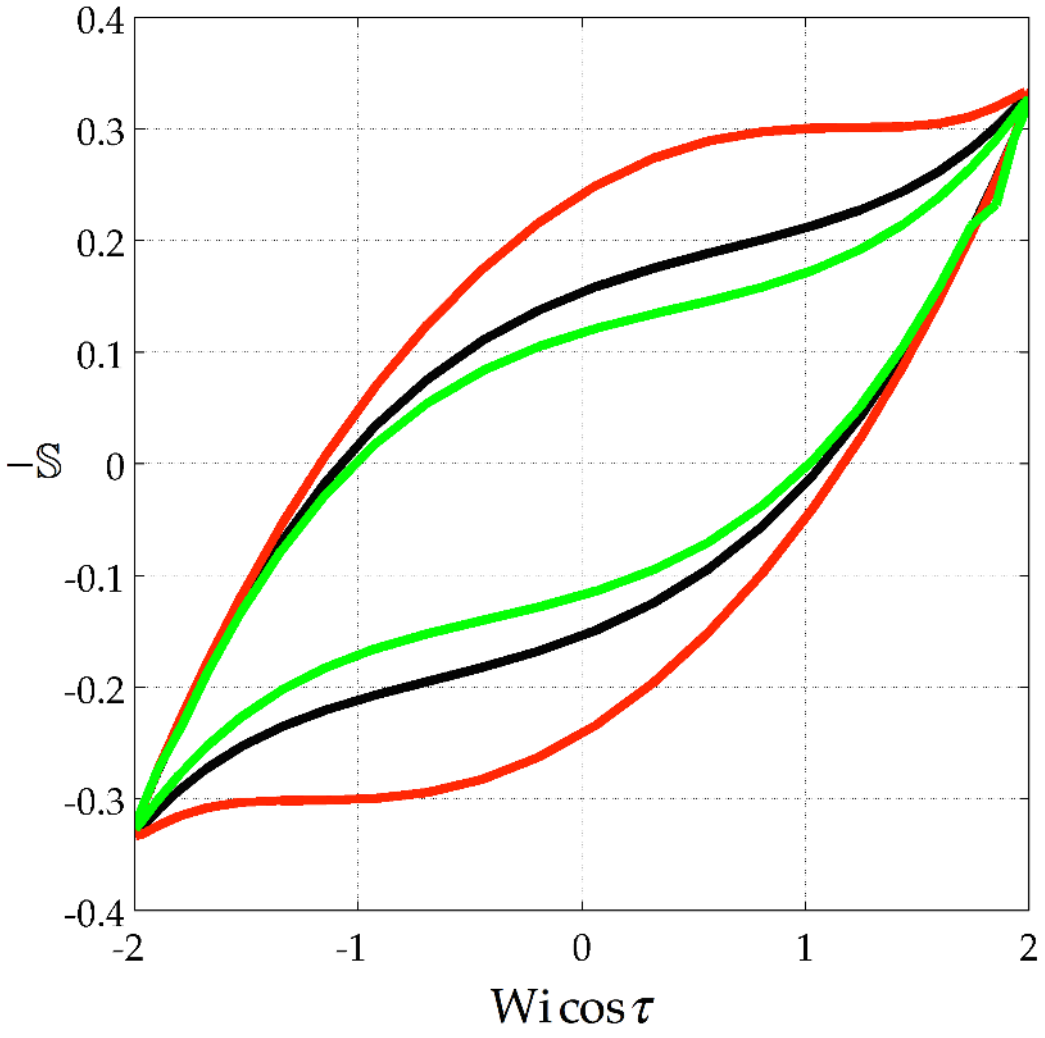

Figure 4. $[2,2]$ Padé approximant of Eq. (28) (**green**) *versus* Eq. (28) (**red**) and also *versus* exact solution [Eq. (21) with Eqs. (22)-(27)] (**black**) for alternance. Counterclockwise loops of minus dimensionless shear stress, $-\mathbb{S}$, *versus* dimensionless shear rate, $\lambda\dot{\gamma}$, calculated for the 2-constant corotational Maxwell model with $\mathrm{Wi}/\mathrm{De} = 2$ and $\lambda\omega = 1$.

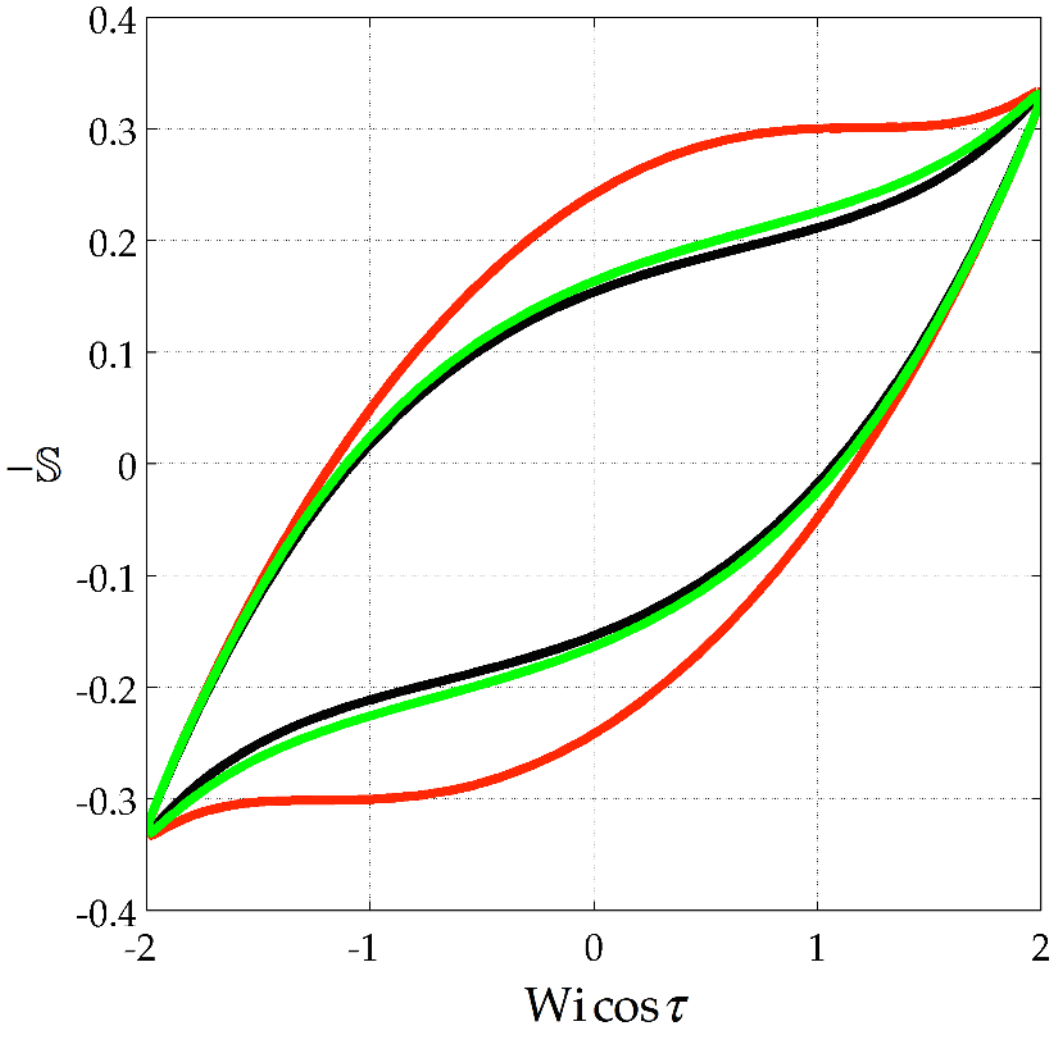

Figure 5. $[2,4]$ Padé approximant of Eq. (28) (**green**) *versus* Eq. (28) (**red**) and also *versus* exact solution [Eq. (21) with Eqs. (22)-(27)] (**black**) for alternance. Counterclockwise loops of minus dimensionless shear stress, $-\mathbb{S}$, *versus* dimensionless shear rate, $\lambda\dot{\gamma}$, calculated for the 2-constant corotational Maxwell model with $\mathrm{Wi}/\mathrm{De} = 2$ and $\lambda\omega = 1$.

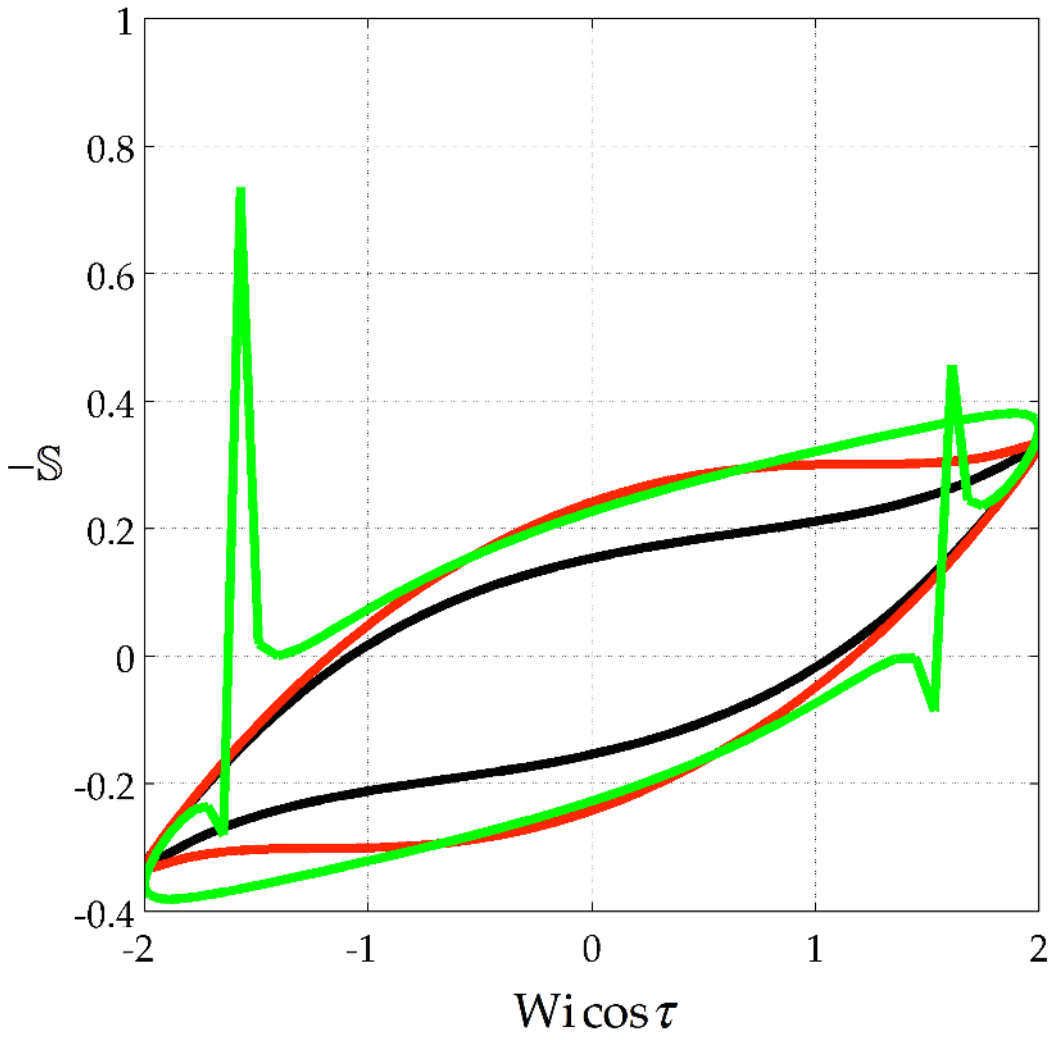

Figure 6. $[0,2]$ Padé approximant of Eq. (39) (**green**) *versus* Eq. (39) (**red**) and also *versus* exact solution [Eq. (21) with Eqs. (22)-(27)] (**black**) for alternance. Counterclockwise loops of minus dimensionless shear stress, $-\mathbb{S}$, *versus* dimensionless shear rate, $\lambda\dot{\gamma}$, calculated for the 2-constant corotational Maxwell model with $\mathrm{Wi}/\mathrm{De} = 2$ and $\lambda\omega = 1$.

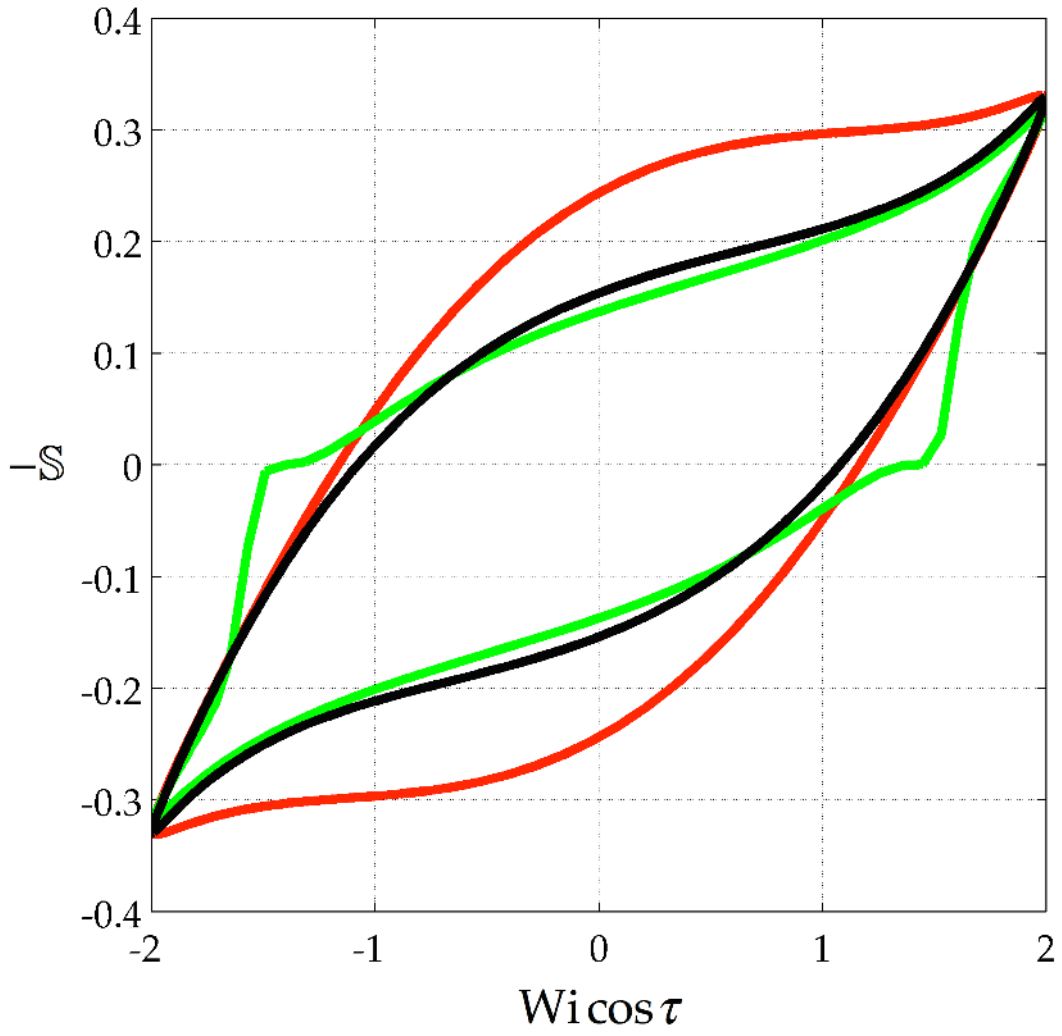

Figure 7. $[0,4]$ Padé approximant of Eq. (39) (**green**) *versus* Eq. (39) (**red**) and also *versus* exact solution [Eq. (21) with Eqs. (22)-(27)] (**black**) for alternance. Counterclockwise loops of minus dimensionless shear stress, $-\mathbb{S}$, *versus* dimensionless shear rate, $\lambda\dot{\gamma}$, calculated for the 2-constant corotational Maxwell model with $\mathrm{Wi}/\mathrm{De} = 2$ and $\lambda\omega = 1$.

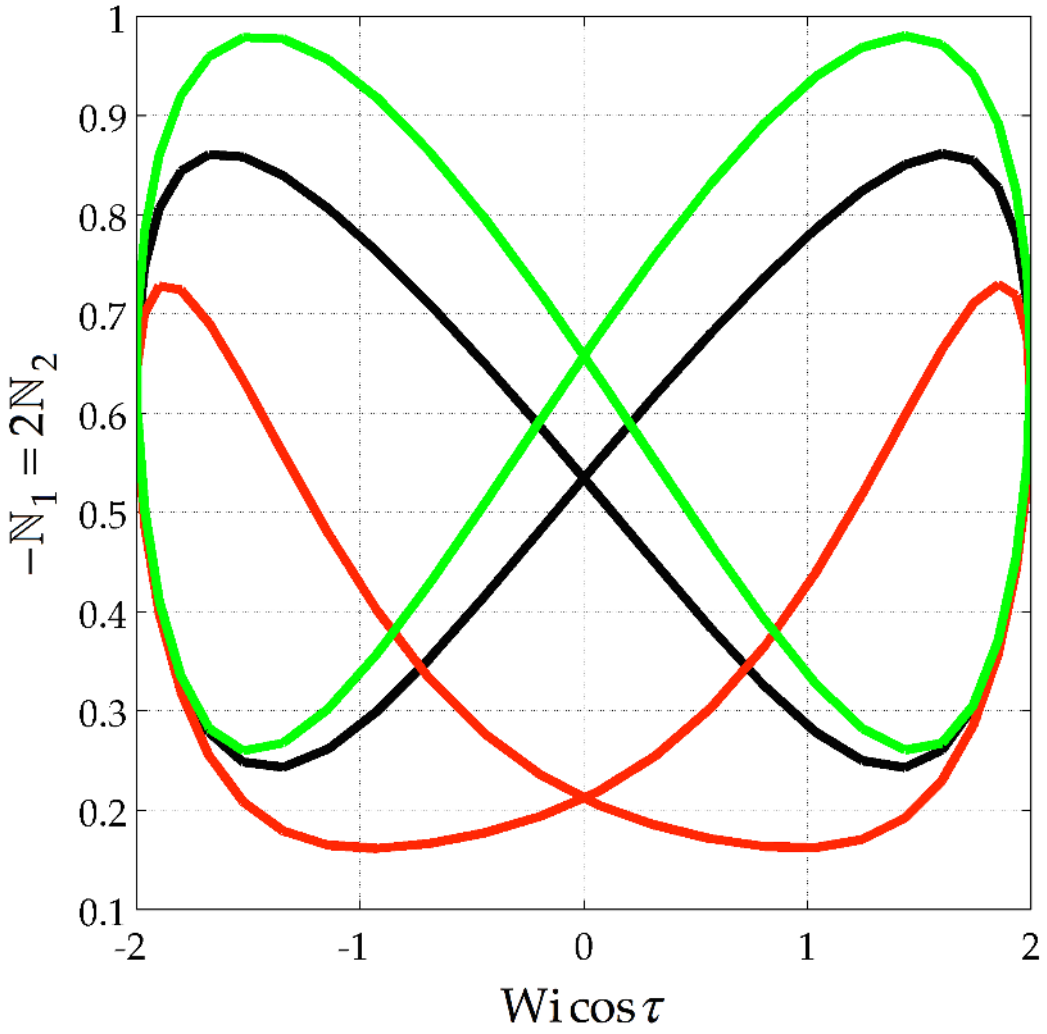

Figure 8. $[1,2]$ Padé approximant of Eq. (42) (**green**) *versus* Eq. (42) (**red**) and also versus exact solution [Eq. (41) with Eqs. (22)-(27)] (**black**) for alternance versus $[x,y]$. Minus dimensionless first normal stress differences, $-\mathbb{N}_1 = 2\mathbb{N}_2$, *versus* dimensionless shear rate, $\lambda\dot{\gamma}$, left-clockwise loops calculated for the 2-constant corotational Maxwell model with $\mathrm{Wi}/\mathrm{De} = 2$ and $\lambda\omega = 1$.

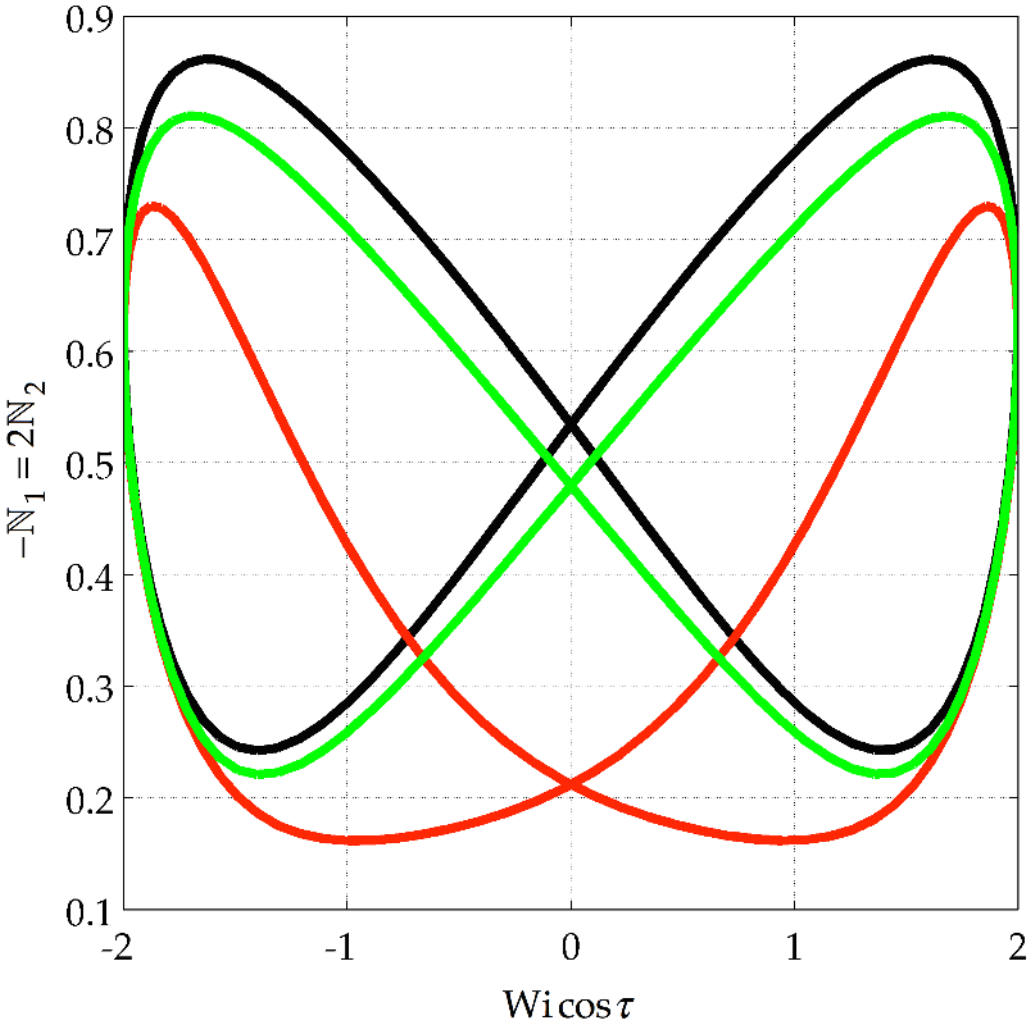

Figure 9. $[1,4]$ Padé approximant of Eq. (42) (**green**) *versus* Eq. (42) (**red**) and also versus exact solution [Eq. (41) with Eqs. (22)-(27)] (**black**) for alternance versus $[x,y]$. Minus dimensionless first normal stress differences, $-\mathbb{N}_1 = 2\mathbb{N}_2$, *versus* dimensionless shear rate, $\lambda\dot{\gamma}$, left-clockwise loops calculated for the 2-constant corotational Maxwell model with $\mathrm{Wi}/\mathrm{De} = 2$ and $\lambda\omega = 1$.

Table I: Literature on Analytical Solutions for Large-Amplitude Oscillatory Shear Flow

	Model	First	Third	Fifth	Zeroth	Second	Fourth	Startup	Notation Eq. (τ_{yx}) (N_1)	Form	[Ref.] (Correction to)
		Shear Stress Harmonic			Normal Stress Difference Harmonic						
Kirkwood and Plock (1956, 1967); Plock (1957)	RD^h, SK^h	n							(12)	$\cong$	[31,32,33]
Lodge (1961, 1964)	L†	ℓ			N_1	N_1				$=$	[34,35]
Spriggs (1966)	NGJ	ℓ			N_1	N_1				$=$	[36]
Spriggs (1966)	CJ	ℓ			N_1 N_2	N_1 N_2				$=$	[36]
Williams and Bird (1962)	O$_3$	ℓ				N_1				$=$	[37]
Williams and Bird (1964)	O$_3$	ℓ			N_1	N_1				$=$	[38]
Spriggs (1965)	O$_3^\dagger$	ℓ			N_1 N_2	N_1 N_2				$=$	[39]
Akers and Williams (1969)	RZ				N_1	N_1				$=$	[40]
Paul (1969); Paul (1970); Bharadwaj (2012)	RD^h, SK^h	n	X						(12)	$\cong$	[41,42,43] (31,33)
Walters and Jones (1970); Walters (1975)	I$_3$	n	X							$\cong$	[44]; Section 6.2.3 of [45]
Paul and Mazo (1969), Paul (1970)	RR^h	n	X						(12)	$\cong$	[46,42]
MacDonald, Marsh and Ashare (1969)	BC, OWFS†	n								$\cong$	[47]
Bird, Warner and Evans (1971)	RD	n			N_1 N_2	N_1 N_2			(12)	$\cong$	[48]
Leal and Hinch (1972)	RSD	ℓ			N_1 N_2	N_1 N_2			(12) (13)	$\cong$	Section 3.3 of [49]
Abdel-Khalik *et al.* (1974); Bird *et al.* (1974)	GE+SK	n			N_1 N_2	N_1 N_2				$\cong$	[50,51]

Bird *et al.* (1977)	BHS	ℓ	0	0						=	Table 11.4-2 [52]
Mou and Mazo (1977)	RR [h]				N_1 N_2	N_1 N_2				≅	[53](46)
Pearson and Rochefort (1982); Helfand and Rochefort (1982)	R [†]	n	X						(11)	≅	[15,16]
Fan and Bird (1984)	CB [†]	n	X						(12)	≅	[18](16)
Oakley (1992); Oakley and Giacomin (1994)	L [†]	ℓ			N_1	N_1				=	APPENDIX B of [54]; [55](34)
Phan-Thien *et al.* (2000)	Dough	ℓ								=	[56]
Yu *et al.* (2002); Zhou (2004)	SE	n	X		N_1	N_1	N_1			≅	[57,58]
Cho *et al.* (2010)	K-BKZ								(11)		[59]
Hoyle (2010)	PP	n							(11)	≅	[60]
Wagner *et al.* (2011)	R	n	X						(11)	≅	[61]
Gurnon and Wagner (2012)	G	ℓ	X		N_1 N_2	N_1 N_2			(11)	≅	[62]
Giacomin *et al.* (2011)	CM [†], CJ	n	X	X	N_1 N_2	N_1 N_2	N_1 N_2	X	(12) (13)	≅	[7](63)
Giacomin and Bird (2011)	ANSR	n	X	X	N_1 N_2	N_1 N_2	N_1 N_2	X	(12) (13)	≅	[64]
Bird *et al.* (2014)	RD	n	X						(12)	≅	[19]
Schmalzer *et al.* (2014)	RD				N_1 N_2	N_1 N_2	N_1 N_2		(13)	≅	[29,30,65]
Saengow *et al.* (2014)	CM	n	X	X	N_1 N_2	N_1 N_2	N_1 N_2	X	(21) (41)	=	[66]
Thompson and de Souza Mendes (2015)	MSJ	n			0	0	0	0		≅	See Section 5.2.2 of [67]
Bozorgi (2014); Bozorgi and Underhill (2014)	AS [h]	n	X						(11)	≅	Chapter 8 of [68]; [69]
This paper	CM	n	X	X	N_1 N_2	N_1 N_2	N_1 N_2		(35) (46)	≅	

Legend: ANSR ≡ corotational; ANSR; AS ≡ active rod suspensions; BC ≡ Bird-Carreau; BHS ≡ Bead-Hookean spring; CB ≡ Curtiss-Bird; CJ ≡ corotational Jeffreys; CM ≡ corotational Maxwell; GE ≡ Goddard integral expansion; L ≡ Lodge rubberlike; NGJ ≡ nonlinear Generalized Jeffreys; O_3 ≡ 3-constant Oldroyd; OWFS ≡ modified Oldroyd-Walters-Fredrickson-Spriggs; PP ≡ pompom; R ≡ reptation; RD ≡ rigid dumbbell; RR ≡ planar rigid ring; RZ ≡ Rouse-Zimm; SE ≡ simple

emulsion; SK ≡ shish-kebab; N_1, N_2 ≡ first and second normal stress differences; $n \equiv \eta^*(\omega, \gamma_0)$; $\ell \equiv \eta^*(\omega)$; † ≡ multiple relaxation times; $=$ ≡ exact; $\cong$ ≡ approximate; ^{h} ≡ with hydrodynamic interaction.

Table II: Dimensional Variables

Angular frequency	t^{-1}	ω
Cartesian coordinate	L	x, y, z
Elastic Fourier modulus	M/Lt^2	G'_n
Extra stress tensor*	M/Lt^2	[redacted]
Extra stress, ijth component	M/Lt^2	τ_{ij}
First normal stress coefficient	ML^{-1}	Ψ_1
First normal stress difference	M/Lt^2	$N_1 \equiv \tau_{11} - \tau_{22}$
First normal stress Fourier coefficient, in phase with $\cos m\tau$, Eq. (13)	$\dfrac{\Psi_1}{\dot{\gamma}^{0^{n-1}}}$	$\Psi'_{1,m,n}$
First normal stress Fourier coefficient, out of phase with $\cos m\tau$, Eq. (13)	$\dfrac{\Psi_1}{\dot{\gamma}^{0^{n-1}}}$	$\Psi''_{1,m,n}$
Fourier coefficients, in phase with $\cos n\tau$, mnth component, Eq. (11)	M/Lt^2	G''_{mn}
Fourier coefficients, out of phase with $\cos n\tau$, mnth component, Eq. (11)	M/Lt^2	G'_{mn}
Loss moduli, in phase with $\cos n\tau$, mnth component, Eq. (12) [70]	M/Lt^2	η'_{mn}
Relaxation time	t	λ
Second normal stress coefficient	ML^{-1}	Ψ_2
Second normal stress difference	M/Lt^2	$N_2 \equiv \tau_{22} - \tau_{33}$
Shear rate, amplitude	t^{-1}	$\dot{\gamma}^0$
Steady shear viscosity	M/Lt	η
Storage moduli, out of phase with $\cos n\tau$, mnth component, Eq. (12) [70]	M/Lt^2	η''_{mn}
Strain rate tensor	t^{-1}	[redacted]
Strain rate, ijth component	t^{-1}	$\dot{\gamma}_{ij}$
Time	t	t
Velocity vector	L/t	$\mathbf{v}$
Velocity, ith-components	L/t	v_i
Viscous Fourier modulus	M/Lt^2	G''_n
Vorticity tensor	t^{-1}	[redacted]
Zero shear rate viscosity	M/Lt	η_0

Legend: M mass; L length; t time

* Where τ_{ij} is the force exerted in the *j*th direction on a unit area of fluid surface of constant x_i by fluid in the region lesser x_i on fluid in the region greater x_i [71].

Table III: Dimensionless Variables and Groups

Bessel function of first kind, mth order	$J_m(z) \equiv \sum_{k=0}^{\infty} \frac{(-1)^k}{(m+k)!k!}\left(\frac{z}{2}\right)^{m+2k}$
Bessel function of first kind, mth order with argument Wi/De	$J_m \equiv J_m(\mathrm{Wi}/\mathrm{De})$
Coefficient, Eq. (24)	$\alpha_k^{(1)}$
Coefficient, Eq. (26)	$\alpha_k^{(2)}$
Coefficient, Eq. (25)	$\beta_k^{(1)}$
Coefficient, Eq. (27)	$\beta_k^{(2)}$
Deborah number	$\mathrm{De} \equiv \lambda\omega$
First normal stress difference	$\mathbb{N}_1 \equiv N_1/\eta_0\dot{\gamma}^0$
Generalized non-Newtonianness	Gn
Inclination of non-Newtonianness	ϕ
Integral, Eq. (22)	I_1
Integral, Eq. (23)	I_2
Normal stress difference expansion coefficients, Eq. (16)	α, β
Second normal stress difference	$\mathbb{N}_2 \equiv N_2/\eta_0\dot{\gamma}^0$
Shear strain amplitude	γ_0
Shear stress	$\mathbb{S} \equiv \tau_{21}/\eta_0\dot{\gamma}^0$
Shear stress expansion coefficients, Eq. (14)	A, B, C
Time	$\tau \equiv \omega t = \mathrm{De}(t/\lambda)$
Weissenberg number	$\mathrm{Wi} \equiv \lambda\dot{\gamma}^0$

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