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**PADÉ APPROXIMANT FOR NORMAL STRESS DIFFERENCES
IN LARGE-AMPLITUDE OSCILLATORY SHEAR FLOW**

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ABSTRACT

Analytical solutions for the normal stress differences in large-amplitude oscillatory shear flow (LAOS), for continuum or molecular models, normally take the form of the first few terms of a series expansion in the shear rate amplitude. Here, we improve the accuracy of these truncated expansions by replacing them with rational functions *called Padé approximants*. Specifically, we examine replacing the truncated expansion for the corotational Jeffreys fluid with its Padé approximants for the normal stress differences. We begin by extending the Goddard integral expansion (GIE) from its third [*J Non-Newt Fluid Mech* **166**, 1081 (2011)] to its fifth order term in the shear rate amplitude. We then explore the Padé approximants for this extension. We uncover the best approximant, the [3,4] approximant, and compare this with both the GIE and the exact solution [*Macromol Theory Simul* **24**, 352 (2015)]. We use Ewoldt grids to show the stunning accuracy of our [3,4] approximant in LAOS. We quantify this accuracy with an objective function and then map its onto Pipkin space. We find the [3,4] approximant to be a simple accurate expression for the normal stress differences in LAOS. Our applications illustrate how to use our new approximant reliably. For this, we use the Spriggs relations to generalize our best approximant to multimode, and then, we compare with measurements on molten high-density polyethylene, and on dissolved polyisobutylene in isobutylene oligomer.

Keywords: Large-amplitude oscillatory shear flow; LAOS; normal stress differences; corotational Jeffreys fluid; Padé approximants; Pipkin space; Goddard-Miller integral expansion; Spriggs relations.

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I. INTRODUCTION

Conceived in 1935 [1,2,3], oscillatory shear flow, when performed at small amplitude, has become the most popular experiment for exploring the physics of elastic liquids. Weissenberg seems to have been first to conceive of LAOS, writing the following in 1948 about oscillatory shear flow. “It is convenient to use the method in two distinct stages*, confining the first stage to ‘infinitesimally’ small amplitudes (...) and leaving the investigation of increasingly larger amplitudes to the second stage (...) in either stage it is necessary to vary the amplitudes at least over one order of magnitude.” The *asterisk* alerts us to the supporting footnote which goes on to say that “The divison [sic] into two stages is particularly useful for thixotropic and other unstable materials, because often it is possible to obtain for them well defined and reproducible results for infinitesimally small amplitudes, even at comparatively high frequencies, while such a possibility vanishes for larger values of amplitudes.” The next contribution to the literature on LAOS comes from Philippoff [4], nearly two decades later, with his use of an analog computer to Fourier analyze the measured shear stress response of a complex fluid in LAOS. Dodge [5] came shortly thereafter with the first use of a digital computer to Fourier analyze the measured shear stress response [6,7], a method that has come to be known as *Fourier transform rheology* (or simply *FT rheology*) [8].

In certain circumstances, functions can be usefully approximated with ratios of polynomials, called rational functions ([9]; see Chapter 1 of [10]). We call each such ratio the $[x,y]$ *Padé approximant*, where x is the numerator order, and y , the denominator order. Curiously, when the starting function is itself a truncated series, one or more of the Padé approximants may approximate the full series more closely than the truncated series from whence it came. Fluid physicists [11,12,13,14,15,16,17,18,19,20,21,22] and, in particular, rheologists [23,24,25,26,27,28,29,30,31,32,33,34] have exploited this useful property of Padé approximants for various purposes. In this paper, we shall exploit this useful property for the prediction or interpretation of the normal stress difference responses of polymeric liquids in large-amplitude oscillatory shear flow (LAOS).

We generate the velocity field in oscillatory shear flow:

$$\mathbf{v} = [v_x, v_y, v_z] = [\dot{\gamma}^0 y \cos \omega t, 0, 0] \quad (1)$$

by confining the fluid to a simple shear apparatus, and then by subjecting one solid-liquid boundary to a coplanar sinusoidal displacement (see this flow field, and the corresponding coordinate system, illustrated in FIG. 1. of [35]). Hence, the corresponding cosinusoidal shear rate is:

$$\dot{\gamma}(t) = \dot{\gamma}^0 \cos \omega t \quad (2)$$

which can be nondimensionalized, using the fluid relaxation time, λ_1 , to:

$$\begin{aligned} \lambda_1 \dot{\gamma}(t) &= \lambda_1 \dot{\gamma}^0 \cos \lambda_1 \omega (t / \lambda_1) \\ &\equiv W \cos De(t / \lambda_1) \end{aligned} \quad (3)$$

in which the Deborah and Weissenberg numbers are given by:

$$\text{De} \equiv \lambda_1 \omega, \quad \text{Wi} \equiv \lambda_1 \dot{\gamma}^0 \quad (4),(5)$$

where De and Wi are each both functions of the fluid and the flow. Whereas De reflects the amount of elasticity, Wi reflects the amount of nonlinearity. In oscillatory shear flow, elastic liquids respond with both a shear stress, and also two normal stress differences (see Fig. 13 of [36] or Fig. 14 of [37]).

For polymeric liquids in oscillatory shear flow, when (see Eq. (11) of [71], Figure 14 of [71]):

$$[\text{Wi} > 1] \cap \left[\frac{\text{Wi}}{\text{De}} > 1 \right] \quad (6)$$

harmonics in the normal stress difference responses, higher than the second ($n > 2$), are commonly observed. Eq. (6) is thus a useful working definition for *large-amplitude oscillatory shear flow* (LAOS).

A few cycles after startup, when the waveforms reach *alternance*, we can then represent these normal stress difference responses with Fourier series:

$$\frac{N_1(\omega, \dot{\gamma}^0, \tau)}{(\dot{\gamma}^0)^2} \equiv \frac{\tau_{xx} - \tau_{yy}}{(\dot{\gamma}^0)^2} = - \sum_{\substack{n=0 \\ \text{even}}}^{\infty} \Psi'_{1,n}(\omega, \dot{\gamma}^0) \cos n\tau + \Psi''_{1,n}(\omega, \dot{\gamma}^0) \sin n\tau \quad (7)$$

$$\frac{N_2(\omega, \dot{\gamma}^0, \tau)}{(\dot{\gamma}^0)^2} \equiv \frac{\tau_{yy} - \tau_{zz}}{(\dot{\gamma}^0)^2} = - \sum_{\substack{n=0 \\ \text{even}}}^{\infty} \Psi'_{2,n}(\omega, \dot{\gamma}^0) \cos n\tau + \Psi''_{2,n}(\omega, \dot{\gamma}^0) \sin n\tau \quad (8)$$

where $\tau \equiv \omega t$. $\Psi_1^d \equiv \Psi'_{1,0}$ and $\Psi_2^d \equiv \Psi'_{2,0}$, called the *displacement* coefficients, reflect time averages of the alternant normal stress difference responses. By contrast, when Eq. (6) does not apply, the normal stress difference responses reduce to (Eq. (64) of [38]; Eqs. (6.39)-(6.44) of [80] and after Ref. 33 of [101]; [39]; APPENDIX B of [92]):

$$\frac{N_1(\omega, \dot{\gamma}^0, \tau)}{(\dot{\gamma}^0)^2} \equiv \frac{\tau_{xx} - \tau_{yy}}{(\dot{\gamma}^0)^2} = - \left[\Psi'_{1,0}(\omega) + \Psi'_{1,2}(\omega) \cos 2\tau + \Psi''_{1,2}(\omega) \sin 2\tau \right] \quad (9)$$

$$\frac{N_2(\omega, \dot{\gamma}^0, \tau)}{(\dot{\gamma}^0)^2} \equiv \frac{\tau_{yy} - \tau_{zz}}{(\dot{\gamma}^0)^2} = - \left[\Psi'_{2,0}(\omega) + \Psi'_{2,2}(\omega) \cos 2\tau + \Psi''_{2,2}(\omega) \sin 2\tau \right] \quad (10)$$

In other words, when Eq. (6) does not apply, harmonics higher than the second, which we call the *higher harmonics*, vanish. Our sign convention for $\Psi''_{1,2}(\omega, \dot{\gamma}^0)$ and $\Psi''_{1,2}(\omega)$ of Eqs. (7) and (9) differs from that of Eq. (40) of [40].

From kinetic molecular theory, with great effort, approximations to the normal stress difference responses in LAOS have been derived for suspensions of rigid dumbbells (Eq. (13) of [100]):

$$\begin{aligned}
\frac{N_1}{(\lambda\dot{\gamma}^0)nkT} = & (\lambda\dot{\gamma}^0) \left\{ -\left[\frac{3}{5(1+W)} \right] - \left[\frac{3(1-2W)}{5(1+4W)(1+W)} \right] \cos 2\omega t - \left[\frac{9}{5(1+4W)(1+W)} \right] \lambda\omega \sin 2\omega t \right\} \\
& + (\lambda\dot{\gamma}^0)^3 \left[\begin{aligned} & \left(\frac{9(21W^2 + 784W + 475)}{350(W+1)^2(4W+1)(9W+25)} \right) \\ & + \left(\frac{3 \begin{pmatrix} -177,820,596W^7 - 1,884,818,889W^6 - 6,101,765,361W^5 \\ -7,119,625,867W^4 - 3,192,891,587W^3 - 688,728,900W^2 \\ +25,370,000W + 19,000,000 \end{pmatrix}}{350(W+1)^3(4W+1)^2(9W+1)(9W+25)(9W+100)(81W+100)} \right) \cos 2\omega t \\ & + \left(\frac{3 \begin{pmatrix} -27,634,932W^7 - 264,300,165W^6 - 431,971,812W^5 \\ +940,723,347W^4 + 2,233,642,922W^3 + 1,364,819,940W^2 \\ +431,340,000W + 50,600,000 \end{pmatrix}}{175(W+1)^3(4W+1)^2(9W+1)(9W+25)(9W+100)(81W+100)} \right) \lambda\omega \sin 2\omega t \\ & + \left(\frac{3(272,160W^4 + 113,121W^3 - 152,944W^2 - 72,261W + 1,900)}{14(W+1)(4W+1)(9W+1)(9W+25)(16W+1)(81W+100)} \right) \cos 4\omega t \\ & + \left(\frac{3(174,960W^4 - 712,611W^3 - 909,195W^2 - 171,664W + 50,600)}{35(W+1)(4W+1)(9W+1)(9W+25)(16W+1)(81W+100)} \right) \lambda\omega \sin 4\omega t \end{aligned} \right] \quad (11)
\end{aligned}$$

and:

$$\begin{aligned}
\frac{N_2}{(\lambda\dot{\gamma}^0)nkT} = & (\lambda\dot{\gamma}^0) \left[-\frac{36W}{35(W+1)(4W+1)} \cos 2\omega t + \frac{12(1-2W)}{35(W+1)(4W+1)} \lambda\omega \sin 2\omega t \right] \\
& + (\lambda\dot{\gamma}^0)^3 \frac{3}{3850} \left[\begin{aligned} & + W \left(\frac{\begin{pmatrix} 676,141,668W^6 + 6,726,036,285W^5 + 22,879,515,861W^4 \\ +30,153,680,741W^3 + 17,670,955,395W^2 + 5,725,307,250W \\ +664,775,000 \end{pmatrix}}{(W+1)^3(4W+1)^2(9W+1)(9W+25)(9W+100)(81W+100)} \right) \cos 2\omega t \\ & + \left(\frac{\begin{pmatrix} 278,553,816W^7 + 3,170,611,026W^6 + 10,488,789,819W^5 \\ +10,645,327,725W^4 + 2,763,261,389W^3 - 250,144,875W^2 \\ -835,447,500W - 157,000,000 \end{pmatrix}}{(W+1)^3(4W+1)^2(9W+1)(9W+25)(9W+100)(81W+100)} \right) \lambda\omega \sin 2\omega t \\ & - \left[\frac{20W(2,241,432W^3 + 2258595W^2 + 330,218W - 361,825) \cos 4\omega t}{(W+1)(4W+1)(9W+1)(9W+25)(16W+1)(81W+100)} \right. \\ & \quad \left. + \frac{200(69,984W^4 - 123,363W^3 - 230,571W^2 - 100,787W + 3,925) \lambda\omega \sin 4\omega t}{(W+1)(4W+1)(9W+1)(9W+25)(16W+1)(81W+100)} \right] \end{aligned} \right] \quad (12)
\end{aligned}$$

where $W \equiv (\lambda\omega)^2$, in which λ is defined by Eq. (6) of [41]. By *great effort*, we mean that the orientation distribution function must first be derived [98,100], before proceeding to Eqs. (11) and (12).

Table I summarizes a list of solutions to constitutive equations in LAOS for the normal stress difference responses, including the classification of Eqs. (11) and (12) in row 17. From columns 2 of Table I, we learn that simple accurate expressions using the method of Padé approximant for the corotational Jeffreys fluid for normal stress differences have yet to be explored. In this work, we will arrive at these by way of Padé approximants for the corotational Jeffreys fluid in LAOS (Section II). Table II defines our dimensional variables, and Table III, the dimensionless ones (which follow Tables 2 and 3 of [97] with adaptations for [42]).

TABLE I of [38] summarizes experimental work on normal stress differences in LAOS (see also Table 1 of [43]; see FIG. 5. and FIG. 6. of [44]). From these tables, we learn that few have undertaken such measurements. To verify our main result, besides the available LAOS measurements on high-density polyethylene (HDPE) melt [92,93], we performed LAOS experiments on 10% by weight solution of polyisobutylene (PIB) in isobutylene oligomer (see Section V). Whereas the HDPE measurements were performed in sliding plate flow with local normal stress transduction [45,46,92,93], the PIB solution measurements were carried out in cone-plate flow with local normal thrust transduction (see Section 8 of [47]).

II. COROTATIONAL JEFFREYS FLUID

The simplest fluid that allows us to explore the role of infinite-shear rate viscosity, η_∞ , on LAOS is the corotational Jeffreys fluid [35,38,71], since $\lambda_2/\lambda_1 = \eta_\infty/\eta_0$ (see Eq. (28) of [35]). This ratio can play an important role in plastics extrusion, as in, for example, the role of the normal stress differences in pipe extrusion ([48,49,50,51,52]). The corotational framework has been closely connected to molecular frameworks [53,54,55]. Also, our work herein on the continuum framework can, by use of the correspondences in Table 1 of [53], also deepen our understanding of how molecular structure intervenes in the normal stress difference responses in LAOS.

The corotational Jeffreys fluid is given by [56,57]:

$$\boldsymbol{\tau} + \lambda_1 \frac{\mathcal{D}\boldsymbol{\tau}}{\mathcal{D}t} = -\eta_0 \left(\dot{\boldsymbol{\gamma}} + \lambda_2 \frac{\mathcal{D}\dot{\boldsymbol{\gamma}}}{\mathcal{D}t} \right) \quad (13)$$

in which:

$$\frac{\mathcal{D}\boldsymbol{\tau}}{\mathcal{D}t} \equiv \frac{D\boldsymbol{\tau}}{Dt} + \frac{1}{2} \{ \boldsymbol{\omega} \cdot \boldsymbol{\tau} - \boldsymbol{\tau} \cdot \boldsymbol{\omega} \}, \quad \dot{\boldsymbol{\gamma}} \equiv \nabla \mathbf{v} + (\nabla \mathbf{v})^\dagger, \quad \boldsymbol{\omega} \equiv \nabla \mathbf{v} - (\nabla \mathbf{v})^\dagger \quad (14), (15), (16)$$

define the Jaumann derivative, rate of strain and vorticity tensors. In Eq. (14), $D\boldsymbol{\tau}/Dt$ is the substantial derivative. We call the derivative given by Eq. (14) *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 [58], and also [59,60,61,62,63].

Recently, we have further explored the Goddard integral expansion (GIE) of the corotational Jeffreys fluid for the normal stress differences by extending to the next term (Wi^5) of GIE (see Eq. (72) of [38]):

$$\mathbb{N}_{1,T} = -2\mathbb{N}_{2,T} = \left(1 - \frac{\lambda_2}{\lambda_1} \right) (A Wi + B Wi^3 + C Wi^5) \quad (17)$$

where:

$$A \equiv - \left(\frac{1}{1 + De^2} + \frac{(1 - 2De^2)\cos 2\tau + 3De\sin 2\tau}{(1 + De^2)(1 + 4De^2)} \right) \quad (18)$$

and:

$$B \equiv \frac{1}{4} \left(\frac{3}{(1+De^2)(1+4De^2)} + \frac{(4-24De^2)\cos 2\tau + 20De\sin 2\tau}{(1+De^2)(1+4De^2)(1+9De^2)} \right. \\ \left. + \frac{(1-35De^2+24De^4)\cos 4\tau + (10-50De^2)De\sin 4\tau}{(1+De^2)(1+4De^2)(1+9De^2)(1+16De^2)} \right) \quad (19)$$

and:

$$C \equiv -\frac{1}{16} \left(\frac{10}{(1+De^2)(1+4De^2)(1+9De^2)} + \frac{15(1-12De^2)\cos 2\tau + 105De\sin 2\tau}{(1+De^2)(1+4De^2)(1+9De^2)(1+16De^2)} \right. \\ \left. + \frac{6(1-71De^2+120De^4)\cos 4\tau + 84(1-11De^2)De\sin 4\tau}{(1+De^2)(1+4De^2)(1+9De^2)(1+16De^2)(1+25De^2)} \right. \\ \left. + \frac{(1-175De^2+1624De^4-720De^6)\cos 6\tau + 21(1-35De^2+84De^4)De\sin 6\tau}{(1+De^2)(1+4De^2)(1+9De^2)(1+16De^2)(1+25De^2)(1+36De^2)} \right) \quad (20)$$

FIG. 2. of [38] shows significant improvement over the one without the Wi^5 term (Eq. (66) of [97]). Eq. (17) reduces to the corotational Maxwell fluid when $\lambda_2 = 0$, and Eq. (17) is also easily extended for an arbitrary normal stress ratio, N_2/N_1 , [43].

The exact solutions for the alternant normal stress difference responses of a corotational Jeffreys fluid in LAOS are (Eq. (69) of [38]):

$$N_1 = -2N_2 = -2e^{\frac{-\tau}{De}} \cos\left(\frac{Wi}{De} \sin \tau\right) \left[\frac{-1}{De} I_1 + \frac{\lambda_2}{\lambda_1} I_3 - \frac{Wi}{De} \frac{\lambda_2}{\lambda_1} I_5 \right] \\ + 2e^{\frac{-\tau}{De}} \sin\left(\frac{Wi}{De} \sin \tau\right) \left[\frac{-1}{De} I_2 + \frac{\lambda_2}{\lambda_1} I_4 + \frac{Wi}{De} \frac{\lambda_2}{\lambda_1} I_6 \right] \quad (21)$$

where $I_1 - I_6$ are defined by (Eqs. (52)-(57) of [38]):

$$I_1 = \frac{4De^2}{Wi} e^{\frac{\tau}{De}} \sum_{k=1}^{\infty} \left[-\frac{2De k^2 J_{2k}}{1+4k^2 De^2} \cos 2k\tau + \frac{k J_{2k}}{1+4k^2 De^2} \sin 2k\tau \right] \quad (22)$$

$$I_2 = \frac{2De^2}{Wi} e^{\frac{\tau}{De}} \sum_{k=1}^{\infty} \left[\frac{(2k-1) J_{2k-1}}{1+(2k-1)^2 De^2} \cos(2k-1)\tau + \frac{De(2k-1)^2 J_{2k-1}}{1+(2k-1)^2 De^2} \sin(2k-1)\tau \right] \quad (23)$$

$$I_3 = De e^{\frac{\tau}{De}} J_1 + De e^{\frac{\tau}{De}} \sum_{k=1}^{\infty} \left[\frac{-J_{2k-1} + J_{2k+1}}{1+4k^2 De^2} \cos 2k\tau + (2De k) \frac{-J_{2k-1} + J_{2k+1}}{1+4k^2 De^2} \sin 2k\tau \right] \quad (24)$$

$$I_4 = De e^{\frac{\tau}{De}} \sum_{k=1}^{\infty} \left[\frac{-(2k-1)De(J_{2k-2} - J_{2k})}{1+(2k-1)^2 De^2} \cos(2k-1)\tau + \frac{J_{2k-2} - J_{2k}}{1+(2k-1)^2 De^2} \sin(2k-1)\tau \right] \quad (25)$$

$$I_5 = \frac{De^2}{Wi} e^{\frac{\tau}{De}} J_1 + De e^{\frac{\tau}{De}} \sum_{k=1}^{\infty} \left[\frac{J_{2k-2} + 2J_{2k} + J_{2k+2}}{2(1+4k^2 De^2)} \cos 2k\tau + (De k) \frac{J_{2k-2} + 2J_{2k} + J_{2k+2}}{1+4k^2 De^2} \sin 2k\tau \right] \quad (26)$$

$$I_6 = \frac{\text{De}}{2} e^{\frac{\tau}{\text{De}}} \sum_{k=1}^{\infty} \left[\frac{-\text{De}(2k-1)(J_{2k-3} + 2J_{2k-1} + J_{2k+1})}{1 + (2k-1)^2 \text{De}^2} \cos(2k-1)\tau \right. \\ \left. + \frac{J_{2k-3} + 2J_{2k-1} + J_{2k+1}}{1 + (2k-1)^2 \text{De}^2} \sin(2k-1)\tau \right] \quad (27)$$

wherein the m th order Bessel function of the first kind of argument Wi/De is given by:

$$J_m\left(\frac{\text{Wi}}{\text{De}}\right) \equiv \sum_{k=0}^{\infty} \frac{(-1)^k}{(m+k)!k!} \left(\frac{\text{Wi}}{2\text{De}}\right)^{m+2k} \quad (28)$$

For brevity, we write J_m to mean $J_m(\text{Wi}/\text{De})$. The existence of both this exact solution (Eq. (21) [with Eqs. (22)-(27)]), and the corresponding GIE (Eq. (17) [with Eqs. (18)-(20)]) presents us with a golden opportunity to see which Padé approximants for the normal stress differences, if any, are accurate. This thus motivates this paper.

For the shear stress in LAOS, we have previously shown that Padé approximants of the GIE, and specifically the [2,4] approximant, is more accurate than the truncated GIE from whence it came [27,34]. In this paper, we assess the usefulness of Padé approximants for improving the accuracy of the truncated GIE for the normal stress difference responses of the corotational Jeffreys fluid in LAOS. We examine all Padé approximants of Eq. (17) [with Eqs. (18)-(20)], following the method of **Section Analysis: corotational Maxwell model. Normal stress differences** of [27], up to order [5,5]. We discover that the [3,4] approximant is accurate, and it is the best of those up to order [5,5]:

$$\mathbb{N}_{1,P} = -2\mathbb{N}_{2,P} = \left(1 - \frac{\lambda_2}{\lambda_1}\right) \frac{(A^2C - AB^2)\text{Wi} + (2ABC - B^3)\text{Wi}^3}{AC - B^2 + BC\text{Wi}^2 - C^2\text{Wi}^4} \quad (29)$$

where A , B and C are defined in Eqs. (18)-(20). Eq. (29) [with Eqs. (18)-(20)] is the main result of this work. By the *best* [x,y], we mean the approximant, over the range $0 \leq \omega t \leq 2\pi$, giving dimensionless normal stress differences *versus* the shear rate loops having one-fold symmetry, and having the lowest value of (x+y). It goes without saying that the main result of this paper overrides the [1,4] approximant from the lower order truncation of GIE, Eq. (46) [with Eqs. (43) and (44)] of [27]. We will use our main result, Eq. (29) [with Eqs. (18)-(20)], to explore its region of accuracy on the Pipkin maps of Section IV. In Section V, we will generalize this main result to multimode using the Spriggs relations. In this very section, we then apply our main result to the LAOS measurements of HDPE (Subsection V.a) and PIB (Subsection V.b).

Figure 1 verifies the steady shear behaviors of our main result, Eq. (29) [with Eqs. (18)-(20)] with the well-known expression for steady shear behaviour (Eq. (67) of [38]). Furthermore, evaluating our main result, Eq. (29) [with Eqs. (18)-(20)] for SAOS, $\text{Wi} \rightarrow 0$, gives:

$$\mathbb{N}_{1,P} = -2\mathbb{N}_{2,P} = 0 \quad (30)$$

that is vanishingly small, as it should (see Eq. 25 of [101]). Furthermore, the

Taylor series expansion of our main result, Eq. (29) [with Eqs. (18)-(20)], about $Wi = 0$, recovers Eq. (64) of [38] for its lowest order term, as it must.

Further to these important consistency checks, we plot frequency-matched Lissajous-Bowditch figures of our main result over the ranges of $0.1 \leq De \leq 10$, $0.10 \leq Wi \leq 1.25$ and $0 \leq \lambda_2/\lambda_1 \leq 1/3$. By *frequency-matched*, we mean that the abscissa frequency matches twice the test frequency. We find that our main result agrees closely with both the GIE from whence it came, Eq. (17) [with Eqs. (18)-(20)], and the exact solution, Eq. (21) [with Eqs. (22)-(27)]. We then test our main result under the extreme conditions, $De = 2$ and $Wi/De = 2$, with the GIE from whence it came, Eq. (17) [with Eqs. (18)-(20)], and also against the exact solution. Figure 2, which demonstrates the significant improvement in accuracy of the Padé approximant of Eq. (17) over the GIE from whence it came.

III. EWOLDT GRIDS

In this paper, we follow Dealy *et al.* (1973) in plotting loops of normal stress differences *versus* $\dot{\gamma}(2\tau) = \dot{\gamma}^0 \cos 2\tau$, since these best bring out material nonlinearities as distortions from ellipticity [64,65,66]. Each such loop is constructed for specific conditions (De, Wi) and then miniaturized and mapped in rows and columns onto Pipkin space, that is, onto a map of Wi *versus* De (see FIG. 2. of [38]). We call this grid of loops an $r \times c$ *Ewoldt grid* (see Fig. 4. and Fig. 8. of [67]; Figures 4-4 through 4-6 in [68]; Fig 1. in [55]; Fig 2 of [34]; Figures 2a and 2b of [69]) where r and c are the numbers of rows and columns. These grids over Pipkin space suggest the region of accuracy for both our main result and its corresponding the GIE.

The general expression for each Ewoldt grid cell is given by:

$$\left[\frac{N_1(De, Wi)}{N_{1,max}} \right]_{i,j} = \frac{N_1(De + 2(i-1), Wi)}{N_{1,max}} + 2(j-1) \quad (31)$$

where $i = 1, 2, \dots, r$ and $j = 1, 2, \dots, c$, and where $i = j = 1$ is the lowest leftmost Ewoldt grid cell, and where $N_{1,max}$ is the peak first normal stress difference given by the exact solution, for each (De, Wi) in LAOS. We use Eq. (17) [with Eqs. (18)-(20)], Eq. (29) [with Eqs. (18)-(20)], and Eq. (21) [with Eqs. (22)-(27)] to create Figure 7 through Figure 10. From these figures, we learn that the ratio λ_2/λ_1 does not affect the frequency-matched loop shapes. Importantly, from these figures, we learn that our main result, the [3,4] approximant for the alternant normal stress difference responses (Eq. (29) [with Eqs. (18)-(20)]), yields singularities when:

$$[Wi > 0.1] \cap \left[\frac{Wi}{De} > 3 \right] \quad (32)$$

and the truncated GIE, [Eq. (17) with Eqs. (18)-(20)], shows unphysical disagreement with the exact solution when:

$$[Wi > 0.1] \cap \left[\frac{Wi}{De} > 2 \right] \quad (33)$$

which fall below Eq. (32). We also learn that within:

$$[Wi \leq 0.1] \cup \left[\frac{Wi}{De} \leq 3 \right] \quad (34)$$

our main result, the [3,4] approximant, can replace the exact solution, Eq. (29) [with Eqs. (18)-(20)]. For completeness, we remark that within:

$$[Wi \leq 0.1] \cup \left[\frac{Wi}{De} \leq 2 \right] \quad (35)$$

the GIE, Eq. (17) [with Eqs. (18)-(20)], and the exact solution, Eq. (29) [with Eqs. (18)-(20)], agrees closely.

IV. OBJECTIVE FUNCTION

We assess the accuracy of our approximate solutions, be they the truncated GIE or its [3,4] Padé approximant. Here, we adapt the objective function for shear stress response in LAOS, defined by Eq. (12) of [34]:

$$\epsilon \equiv \sqrt{\oint (\tilde{N}_i - N_i)^2 / N_{i,\max}^2 d\tau} \quad (36)$$

in which, $i = 1, 2$, where $\tilde{N}_i$ is approximate, and, N_i , exact. For the corotational Jeffreys fluid, where $N_1 = -2N_2$, we can first apply Eq. (36) to the truncated GIE [Eq. (17), with Eqs. (18)-(20)] to give:

$$\epsilon_T \equiv \sqrt{\oint (N_{1,T} - N_1)^2 / N_{1,\max}^2 d\tau} = \frac{1}{N_{1,\max}} \sqrt{\int_0^{2\pi} (N_{1,T} - N_1)^2 d\tau} \quad (37)$$

and, next, to our [3,4] Padé approximant of Eq. (17) [Eq. (29), with Eqs. (18)-(20)]:

$$\epsilon_P \equiv \sqrt{\oint (N_{1,P} - N_1)^2 / N_{1,\max}^2 d\tau} = \frac{1}{N_{1,\max}} \sqrt{\int_0^{2\pi} (N_{1,P} - N_1)^2 d\tau} \quad (38)$$

We thus quantify inaccuracies with the objective functions, ϵ_T and ϵ_P , and then map these onto Pipkin space. In Eq. (37), $N_{1,T}$ is given by Eq. (17), and in Eq. (38), $N_{1,P}$ by Eq. (29). In both Eqs. (37) and (38), N_1 is given by Eq. (21) [with Eqs. (22)-(27)]. We use Eq. (38) [with Eqs. (21) and (17)], to plot Figure 11, and Eq. (37) [with Eqs. (21) and (29)] to plot Figure 12.

Comparing ϵ_P and ϵ_T (Figure 11 *versus* Figure 12), we see that our main result, the [3,4] Padé approximant [Eq. (29) with Eqs. (18)-(20)] is much more accurate than the truncated GIE from whence it came. Whereas we find our [3,4] Padé approximant to be useful when:

$$[Wi \leq 0.1] \cup \left[\frac{Wi}{De} \leq 3 \right] \quad (39)$$

the truncated GIE is only useful when:

$$[\text{Wi} \leq 0.1] \cup \left[\frac{\text{Wi}}{\text{De}} \leq 2 \right] \quad (40)$$

From Figure 11 and Figure 12, we learn that the truncated GIE always departs from the exact solution before our [3,4] Padé approximant, and we also learn that increasing De always improves the accuracy of our main result [Eq. (29) with Eqs. (18)-(20)].

V. APPLICATIONS: POLYMERIC LIQUIDS

In this section, we apply our main result, Eq. (29) [with Eqs. (18)-(20)], to the measured LAOS behaviors of molten high-density polyethylene (HDPE) and of dissolved polyisobutylene (PIB). We first *generalize* both our main result, Eq. (29) [with Eqs. (18)-(20)], and the corresponding GIE, Eq. (17) [with Eqs. (18)-(20)], to multiple relaxation and retardation times. We then fit η_0 , λ_1 , λ_2 and the Spriggs exponent, α , to the SAOS measurements, $\eta'(\omega)$ and $\eta''(\omega)$. Finally, we test our main result against the measured LAOS normal stress difference behaviors.

Spriggs relations have been used successfully to generalize constitutive equations for polymeric liquids to multimode analytically [34,70, 71,82,85,101]. The Spriggs relations discretize the polymeric system into an infinity of relaxation and retardation times as follows (Eqs. (6.1-14) and (6.1-15) of [58]), and Eq. (182) of [70]:

$$\eta_j = \eta_0 \lambda_{1,j} / \sum_{j=1}^{\infty} \lambda_{1,j}, \quad \lambda_{1,j} = \lambda_1 / j^\alpha, \quad \lambda_{2,j} = \lambda_2 / j^\alpha \quad (41),(42),(43)$$

For the corotational Jeffreys fluid, we then get (see Eqs. (69) and (70) of [71]):

$$\frac{\eta'(\omega)}{\eta_0} = \frac{1}{\zeta(\alpha)} \sum_{j=1}^{\infty} \frac{j^\alpha + \lambda_1 \lambda_2 j^{-\alpha} \omega^2}{j^{2\alpha} + \lambda_1^2 \omega^2}, \quad \frac{\eta''(\omega)}{\eta_0} = \frac{1}{\zeta(\alpha)} \sum_{j=1}^{\infty} \frac{(\lambda_1 - \lambda_2) \omega}{j^{2\alpha} + \lambda_1^2 \omega^2} \quad (44),(45)$$

in which $\zeta(\alpha) \equiv \sum_{j=1}^{\infty} j^{-\alpha}$ is the Riemann zeta function.

We next use Eqs. (41)-(43) to generalize the exact solution, Eq. (21), to get:

$$\sum_{j=1}^{\infty} \mathbb{N}_1^{(j)} = \sum_{j=1}^{\infty} \frac{N_1^{(j)}}{\eta_0 \dot{\gamma}^0} = \frac{2}{\eta_0} \sum_{j=1}^{\infty} \eta_j e^{\frac{-\tau}{\lambda_{1,j} \omega}} \left(-\cos\left(\frac{\dot{\gamma}^0}{\omega} \sin \tau\right) \left[\frac{-1}{\lambda_{1,j} \omega} I_{1,j} + \frac{\lambda_{2,j}}{\lambda_{1,j}} I_{3,j} - \frac{\dot{\gamma}^0}{\omega} \frac{\lambda_{2,j}}{\lambda_{1,j}} I_{5,j} \right] + \sin\left(\frac{\dot{\gamma}^0}{\omega} \sin \tau\right) \left[\frac{-1}{\lambda_{1,j} \omega} I_{2,j} + \frac{\lambda_{2,j}}{\lambda_{1,j}} I_{4,j} + \frac{\dot{\gamma}^0}{\omega} \frac{\lambda_{2,j}}{\lambda_{1,j}} I_{6,j} \right] \right) \quad (46)$$

where $I_{1,j} - I_{6,j}$ are defined by:

$$I_{1,j} = \frac{4(\lambda_{1,j} \omega)^2}{\lambda_{1,j} \dot{\gamma}^0} e^{\frac{-\tau}{\lambda_{1,j} \omega}} \sum_{k=1}^{\infty} \left[-\frac{2(\lambda_{1,j} \omega) k^2 J_{2k}}{1 + 4k^2 (\lambda_{1,j} \omega)^2} \cos 2k\tau + \frac{k J_{2k}}{1 + 4k^2 (\lambda_{1,j} \omega)^2} \sin 2k\tau \right] \quad (47)$$

$$I_{2,j} = \frac{2(\lambda_{1,j}\omega)^2}{\lambda_{1,j}\dot{\gamma}^0} e^{\frac{\tau}{\lambda_{1,j}\dot{\gamma}^0}} \sum_{k=1}^{\infty} \left[\frac{(2k-1)J_{2k-1}}{1+(2k-1)^2(\lambda_{1,j}\omega)^2} \cos(2k-1)\tau \right. \\ \left. + \frac{\lambda_{1,j}\omega(2k-1)^2 J_{2k-1}}{1+(2k-1)^2(\lambda_{1,j}\omega)^2} \sin(2k-1)\tau \right] \quad (48)$$

$$I_{3,j} = \lambda_{1,j}\omega e^{\frac{\tau}{\lambda_{1,j}\omega}} J_1 + \lambda_{1,j}\omega e^{\frac{\tau}{\lambda_{1,j}\omega}} \sum_{k=1}^{\infty} \left[\frac{-J_{2k-1} + J_{2k+1}}{1+4k^2(\lambda_{1,j}\omega)^2} \cos 2k\tau \right. \\ \left. + (2\lambda_{1,j}\omega k) \frac{-J_{2k-1} + J_{2k+1}}{1+4k^2(\lambda_{1,j}\omega)^2} \sin 2k\tau \right] \quad (49)$$

$$I_{4,j} = \lambda_{1,j}\omega e^{\frac{\tau}{\lambda_{1,j}\omega}} \sum_{k=1}^{\infty} \left[\frac{-(2k-1)\lambda_{1,j}\omega(J_{2k-2} - J_{2k})}{1+(2k-1)^2(\lambda_{1,j}\omega)^2} \cos(2k-1)\tau \right. \\ \left. + \frac{J_{2k-2} - J_{2k}}{1+(2k-1)^2(\lambda_{1,j}\omega)^2} \sin(2k-1)\tau \right] \quad (50)$$

$$I_{5,j} = \frac{(\lambda_{1,j}\omega)^2}{\lambda_{1,j}\dot{\gamma}^0} e^{\frac{\tau}{\lambda_{1,j}\omega}} J_1 + \lambda_{1,j}\omega e^{\frac{\tau}{\lambda_{1,j}\omega}} \sum_{k=1}^{\infty} \left[\frac{J_{2k-2} + 2J_{2k} + J_{2k+2}}{2(1+4k^2(\lambda_{1,j}\omega)^2)} \cos 2k\tau + (\lambda_{1,j}\omega k) \frac{J_{2k-2} + 2J_{2k} + J_{2k+2}}{1+4k^2(\lambda_{1,j}\omega)^2} \sin 2k\tau \right] \quad (51)$$

$$I_{6,j} = \frac{\lambda_{1,j}\omega}{2} e^{\frac{\tau}{\lambda_{1,j}\omega}} \sum_{k=1}^{\infty} \left[\frac{-\lambda_{1,j}\omega(2k-1)(J_{2k-3} + 2J_{2k-1} + J_{2k+1})}{1+(2k-1)^2(\lambda_{1,j}\omega)^2} \cos(2k-1)\tau \right. \\ \left. + \frac{J_{2k-3} + 2J_{2k-1} + J_{2k+1}}{1+(2k-1)^2(\lambda_{1,j}\omega)^2} \sin(2k-1)\tau \right] \quad (52)$$

We next use Eqs. (41)-(43) to generalize the truncated GIE solution for normal stress difference, Eq. (17), to multimode:

$$\sum_{j=1}^{\infty} \mathbb{N}_{1,T}^{(j)} = \sum_{j=1}^{\infty} \frac{N_{1,T}^{(j)}}{\eta_0 \dot{\gamma}^0} = \frac{1}{\eta_0} \sum_{j=1}^{\infty} \eta_j \left(1 - \frac{\lambda_{2,j}}{\lambda_{1,j}} \right) \left(A_j \lambda_{1,j} \dot{\gamma}^0 + B_j (\lambda_{1,j} \dot{\gamma}^0)^3 + C_j (\lambda_{1,j} \dot{\gamma}^0)^5 \right) \quad (53)$$

$$A_j = - \left(\frac{1}{1 + \lambda_{1,j}^2 \omega^2} + \frac{(1 - 2\lambda_{1,j}^2 \omega^2) \cos 2\tau + 3\lambda_{1,j} \omega \sin 2\tau}{(1 + \lambda_{1,j}^2 \omega^2)(1 + 4\lambda_{1,j}^2 \omega^2)} \right) \quad (54)$$

and:

$$B_j = \frac{1}{4} \left(\frac{3}{(1 + \lambda_{1,j}^2 \omega^2)(1 + 4\lambda_{1,j}^2 \omega^2)} \right. \\ \left. + \frac{(4 - 24\lambda_{1,j}^2 \omega^2) \cos 2\tau + 20\lambda_{1,j} \omega \sin 2\tau}{(1 + \lambda_{1,j}^2 \omega^2)(1 + 4\lambda_{1,j}^2 \omega^2)(1 + 9\lambda_{1,j}^2 \omega^2)} \right. \\ \left. + \frac{(1 - 35\lambda_{1,j}^2 \omega^2 + 24\lambda_{1,j}^4 \omega^4) \cos 4\tau + (10 - 50\lambda_{1,j}^2 \omega^2) \lambda_{1,j} \omega \sin 4\tau}{(1 + \lambda_{1,j}^2 \omega^2)(1 + 4\lambda_{1,j}^2 \omega^2)(1 + 9\lambda_{1,j}^2 \omega^2)(1 + 16\lambda_{1,j}^2 \omega^2)} \right) \quad (55)$$

and:

$$C_j = -\frac{1}{16} \left[\begin{aligned} & \frac{10}{(1+\lambda_{1,j}^2\omega^2)(1+4\lambda_{1,j}^2\omega^2)(1+9\lambda_{1,j}^2\omega^2)} \\ & + \frac{15(1-12\lambda_{1,j}^2\omega^2)\cos 2\tau + 105\lambda_{1,j}\omega\sin 2\tau}{(1+\lambda_{1,j}^2\omega^2)(1+4\lambda_{1,j}^2\omega^2)(1+9\lambda_{1,j}^2\omega^2)(1+16\lambda_{1,j}^2\omega^2)} \\ & + \frac{6(1-71\lambda_{1,j}^2\omega^2+120\lambda_{1,j}^4\omega^4)\cos 4\tau + 84(1-11\lambda_{1,j}^2\omega^2)\lambda_{1,j}\omega\sin 4\tau}{(1+\lambda_{1,j}^2\omega^2)(1+4\lambda_{1,j}^2\omega^2)(1+9\lambda_{1,j}^2\omega^2)(1+16\lambda_{1,j}^2\omega^2)(1+25\lambda_{1,j}^2\omega^2)} \\ & + \frac{(1-175\lambda_{1,j}^2\omega^2+1624\lambda_{1,j}^4\omega^4-720\lambda_{1,j}^6\omega^6)\cos 6\tau + 21(1-35\lambda_{1,j}^2\omega^2+84\lambda_{1,j}^4\omega^4)\lambda_{1,j}\omega\sin 6\tau}{(1+\lambda_{1,j}^2\omega^2)(1+4\lambda_{1,j}^2\omega^2)(1+9\lambda_{1,j}^2\omega^2)(1+16\lambda_{1,j}^2\omega^2)(1+25\lambda_{1,j}^2\omega^2)(1+36\lambda_{1,j}^2\omega^2)} \end{aligned} \right] \quad (56)$$

We then use Eqs. (41)-(43) to generalize our [3,4] Padé approximant for the normal stress differences, Eq. (29), to multimode:

$$\sum_{j=1}^{\infty} N_{1,P}^{(j)} = \sum_{j=1}^{\infty} \frac{N_{1,P}^{(j)}}{\eta_0 \dot{\gamma}^0} = \frac{1}{\eta_0} \sum_{j=1}^{\infty} \eta_j \left(1 - \frac{\lambda_{2,j}}{\lambda_{1,j}} \right) \frac{(A_j^2 C_j - A_j B_j^2) \lambda_{1,j} \dot{\gamma}^0 + (2A_j B_j C_j - B_j^3) (\lambda_{1,j} \dot{\gamma}^0)^3}{A_j C_j - B_j^2 + B_j C_j (\lambda_{1,j} \dot{\gamma}^0)^2 - C_j^2 (\lambda_{1,j} \dot{\gamma}^0)^4} \quad (57)$$

where A_j , B_j and C_j are given by Eqs. (54)-(56).

a. Molten High-Density Polyethylene (HDPE)

A commercial pipe extrusion grade of HDPE (Phillips TR480) containing 2% by weight of carbon black was characterized for N_1 in LAOS at 170°C by sliding plate rheometry incorporating local pressure transduction (Section III of [93] and Section IV.5.2 of [92], [45]). The SAOS measurements of $\eta'(\omega)$ and $\eta''(\omega)$ for this material have also been previously reported (see Section II.4 of [92]). We begin by fitting Eqs. (44) and (45) to the $\eta'(\omega)$ and $\eta''(\omega)$ measurements (see Figure 10. and Figure 11. of [92]) to get $\eta_0 = 0.180 \text{ MPa} \cdot \text{s}$, $\lambda_1 = 60.0 \text{ s}$, $\lambda_2 = 0.500 \text{ s}$ and $\alpha = 2.31$. Figure 13 shows the goodness of fit. Finally, we test our generalized main result, Eq. (57) [with Eqs. (54)-(56)], against LAOS measurements (see Figure 112. of [92]). Figure 14 shows good agreement for the displacement coefficient, but the loop areas for the HDPE are overpredicted. In other words, the $\Psi_{1,2}''$ is overpredicted (see after Eq. (8) of [38]).

b. Dissolved Polyisobutylene (PIB)

The 10% by weight solution of PIB dissolved in isobutylene oligomer was characterized at 20.4°C by MCR 702 rheometer in cone-plate flow with plate partitioning [72,73,74,75,76,77]. See Section 4.4.3 of [47] for the SAOS measurements, and Section 8.3.1 of [47], for the LAOS. We begin by fitting Eqs. (44) and (45) to the $\eta'(\omega)$ and $\eta''(\omega)$ measurements (see Figure 4.14 of [47]) to get $\eta_0 = 30.0 \text{ Pa} \cdot \text{s}$, $\lambda_1 = 0.470 \text{ s}$, $\lambda_2 = 5.00 \text{ ms}$ and $\alpha = 1.40$. Figure 15 shows the goodness of fit. Finally, we test our generalized main result, Eq. (57) [with Eqs. (54)-(56)], against our LAOS measurements. Figure 16 shows

good agreement between our new [3,4] Padé approximant and the measured behavior of the first normal stress difference of the PIB solution. Slight overprediction of the displacement term, $\Psi_1^d(\omega, \dot{\gamma}^0)$, is observed.

VI. CONCLUSION

In this paper, we focus on truncated expansions for the normal stress difference responses of polymeric liquids subjected to LAOS. We use the corotational Jeffreys fluid for which both an exact solution and a truncated Goddard integral expansion (up to Wi^5) are available. We discover the [3,4] Padé approximant with which such truncated expansions can be replaced, with much improved accuracy. This [3,4] Padé approximant (Eq. (29) [with Eqs. (18)-(20)]) is thus more accurate than the truncated expansion for the normal stress difference response from whence the approximant came.

We compare our main result with experimental LAOS measurements for molten HDPE and for dissolved PIB. For this comparison, we generalize our corotational Jeffreys fluid to multimode using the Spriggs relations. Comparing our generalized main result with the LAOS measurements of molten HDPE shows good agreement in the displacement term, $\Psi_1^d(\omega, \dot{\gamma}^0)$, but $\Psi_{1,2}''(\omega, \dot{\gamma}^0)$ is overpredicted. For the dissolved PIB solution, our generalized main result for the corotational Jeffrey fluid agrees with our new LAOS measurements. We know of no measurement of the second normal stress difference response to LAOS, though this has been measured in SAOS [78, 79, 43].

Thus for truncated expansions made available from molecular theory, such as those given in Eqs. (11) and (12) for suspensions of rigid dumbbells, we can recommend the use of Eq. (29) [with Eqs. (18)-(20)] to improve accuracy over the range given by Eq. (39). Of course, from molecular theory, we proceed without the luxury of exact solutions. This thus highlights the usefulness of our new [3,4] approximant to molecular theorists.

Some find the evaluation of the Bessel functions in the exact solution Eq. (21) [with Eqs. (22)-(27)] to be cumbersome. Where Eq. (39) subtends, our new [3,4] approximant altogether obviates Bessel functions.

VII. ACKNOWLEDGMENT

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Table I: Literature on Analytical Solutions for Normal Stress Differences in Large-Amplitude Oscillatory Shear Flow

Authors (year)	Model	Harmonic				Orientation	Startup	Form	References
		Zeroth	Second	Fourth	Sixth				
Lodge (1961, 1964)	$L^{\dagger}$	I	I	0				=	[80,81]
Spriggs (1966)	NGJ	I	I					=	[82]
Spriggs (1966)	CJ	I,II	I,II					=	[82]
Williams and Bird (1962)	O_3		I					=	[83]
Williams and Bird (1964)	O_3	I	I					=	[84]
Spriggs (1965)	$O_3^{\dagger}$	I,II	I,II					=	[85]
Akers and Williams (1969)	RZ	I	I					=	[86]
Bird, Warner and Evans (1971)	RD	I,II	I,II			ψ		$\cong$	[87]
Leal and Hinch (1972)	RSD	I,II	I,II			ψ		$\cong$	Section 3.3 of [88]
Abdel-Khalik <i>et al.</i> (1974); Bird <i>et al.</i> (1974)	GE+SK	I,II	I,II					$\cong$	[89,90]
Mou and Mazo (1977)	RR^h	I,II	I,II			ψ		$\cong$	[91]
Oakley (1992); Oakley and Giacomin (1994)	$L^{\dagger}$	I	I					=	APPENDIX B of [92]; [93]
Yu <i>et al.</i> (2002); Zhou (2004)	SE	I	I	I				$\cong$	[94,95]
Gurnon and Wagner (2012)	G	I,II	I,II					$\cong$	[96]
Giacomin <i>et al.</i> (2011)	$CM^{\dagger}, CJ$	I,II	I,II	I,II			X	$\cong$	[97]
Giacomin and Bird (2011)	ANSR	I,II	I,II	I,II			X	$\cong$	[43]
Schmalzer (2014); Schmalzer <i>et al.</i> (2015);	RD	I,II	I,II	I,II		ψ		$\cong$	[98,99,100]
Saengow <i>et al.</i> (2015)	CM	I,II	I,II	I,II			X	=	[101]
Thompson and de Souza Mendes (2015)	MSJ	0	0	0			0	$\cong$	See Section 5.2.2 of [102]
Giacomin <i>et al.</i> (2015)	CM^P	I,II	I,II	I,II				$\cong$	[27]
Saengow and Giacomin (2017)	O8	I,II	I,II	I,II	I,II			=	[38]
This paper	$CJ^{P,\dagger}$	I,II	I,II	I,II	I,II			$\cong$	[103]

Legend: ANSR $\equiv$ corotational arbitrary normal stress ratio; CJ $\equiv$ corotational Jeffreys; CM $\equiv$ corotational Maxwell; GE $\equiv$ Goddard integral expansion; L $\equiv$ Lodge rubberlike; NGJ $\equiv$ nonlinear Generalized Jeffreys; O_3 $\equiv$ 3-constant Oldroyd; P $\equiv$ Padé Approximant; RD $\equiv$ rigid dumbbell; RR $\equiv$ planar rigid ring; RZ $\equiv$ Rouse-Zimm; SE $\equiv$ simple emulsion; SK $\equiv$ shish-kebab; W $\equiv$ Wagner E I,II $\equiv$ first and second normal stress differences; $\dagger$ $\equiv$ multiple relaxation times; = $\equiv$ exact; $\cong$ $\equiv$ approximate; h $\equiv$ with hydrodynamic interaction; ψ $\equiv$ orientation distribution

Table II: Dimensional Variables

Name	Unit	Variable
Angular frequency [Eq. (2)]	t^{-1}	ω
Cartesian coordinate	L	x, y, z
Velocity vector [Eq. (1)]	L/t	$\mathbf{v}$
Strain rate [Eq. (2)]	t^{-1}	$\dot{\gamma}$
Extra stress tensor* [Eq. (13)]	M/Lt^2	$\boldsymbol{\tau}$
Vorticity tensor [Eq. (14)]	t^{-1}	$\boldsymbol{\omega}$
Rate of strain tensor [Eq. (15)]	t^{-1}	$\dot{\boldsymbol{\gamma}}$
First normal stress coefficient, in phase and out of phase with shear rate, n th harmonic [Eq. (7)]	M/L	$\Psi'_{1,n}, \Psi''_{1,n}$
Second normal stress coefficient, in phase and out of phase with shear rate, n th harmonic [Eq. (8)]	M/L	$\Psi'_{2,n}, \Psi''_{2,n}$
First normal stress coefficients, small amplitude oscillatory shear response, distance term [Eq. (7)]	M/L	$\Psi_1^d \equiv \Psi'_{1,0}$
Second normal stress coefficients, small amplitude oscillatory shear response, distance term [Eq. (8)]	M/L	$\Psi_2^d \equiv \Psi'_{2,0}$
First normal stress difference [Eq. (7)]	M/Lt^2	$N_1 \equiv \tau_{xx} - \tau_{yy}$
Second normal stress difference [Eq. (8)]	M/Lt^2	$N_2 \equiv \tau_{yy} - \tau_{zz}$
First normal stress difference, the peak [Eq. (31)]	M/Lt^2	$N_{1,\max}$
Shear rate, amplitude [Eq. (2)]	t^{-1}	$\dot{\gamma}^0$
Loss [Eq. (44)] and storage [Eq. (45)] viscosity	M/Lt^2	η', η''
Strain rate [Eq. (2)]	t^{-1}	$\dot{\gamma}$
Velocity vector [Eq. (1)]	L/t	$\mathbf{v}$
Relaxation and retardation times [Eq. (3)]	t	λ_1, λ_2
Relaxation time [Eq. (11)]	t	λ
Boltzmann constant [Eq. (11)]	$\frac{M/L^2 t^2 T}{\text{dumbell}}$	k
Viscosity, zero [Eq. (13)] and infinite shear rates [before Eq. (13)]	M/Lt	η_0, η_∞
First normal stress difference, exact solution j th spectrum [Eq. (46)]	M/Lt^2	$N_1^{(j)}$
First normal stress difference, truncated GIE j th spectrum [Eq. (53)]	M/Lt^2	$N_{1,T}^{(j)}$
First normal stress difference, [3,4] Padé approximant j th spectrum [Eq. (57)]	M/Lt^2	$N_{1,P}^{(j)}$
Molar concentration of dumbbells/ chains [Eq. (11)]	$\text{dumbbells}/L^3$	n

Legend: $M \equiv$ mass; $L \equiv$ length; $t \equiv$ time; $T \equiv$ temperature

* 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 (see “Note on the Sign Convention for the Stress Tensor” on pp. 19-20 of [104], or pp. 24-25 of [105]).

Table III: Dimensionless Variables and Groups

Name	Symbol
Deborah number [Eq. (4)]	$De \equiv \lambda_1 \omega$
Deborah number squared [Eq. (11)]	$W \equiv (\lambda \omega)^2$
Functions in Eqs. (18)-(20)	A, B, C
Functions in Eqs. (54)-(56), j th spectrum of those defined in Eqs. (18)-(20)	A_j, B_j, C_j
Functions in Eqs. (22)-(27)	$I_1, \dots, I_6$
Functions in Eqs. (47)-(52), j th spectrum of those defined in Eqs. (22)-(27)	$I_{1,j}, \dots, I_{6,j}$
Degree of Padé approximant, [numerator, denominator]	$[x, y]$
First normal stress difference: exact [Eq. (21)], truncated GIE [Eq. (17)], and [3,4] Padé approximant [Eq. (29)] solutions	$\mathbb{N}_1 = N_1 / \eta_0 \dot{\gamma}^0,$ $\mathbb{N}_{1,P} = N_{1,P} / \eta_0 \dot{\gamma}^0,$ $\mathbb{N}_{1,T} = N_{1,T} / \eta_0 \dot{\gamma}^0$
Second normal stress difference: exact [Eq. (21)], truncated GIE [Eq. (17)], and [3,4] Padé approximant [Eq. (29)] solutions	$\mathbb{N}_2 = N_2 / \eta_0 \dot{\gamma}^0,$ $\mathbb{N}_{2,P} = N_{2,P} / \eta_0 \dot{\gamma}^0,$ $\mathbb{N}_{2,T} = N_{2,T} / \eta_0 \dot{\gamma}^0$
Normal stress differences, exact solution, the peak [Eqs. (31)]	$\mathbb{N}_{1,\max}$
Error [Eq. (36)]	ϵ
Error, GIE expansion approximation [Eq. (37)] and Padé approximant [Eq. (38)]	ϵ_T, ϵ_P
Normal stress difference, an exact solution [Eq. (36)]	$\mathbb{N}_i$
Normal stress difference, an approximation solution [Eq. (36)]	$\tilde{\mathbb{N}}_i$
Normal stress difference, an exact solution, the peak [Eq. (36)]	$\mathbb{N}_{i,\max}$
First normal stress difference, exact solution [Eq. (46)], [3,4] Padé approximant [Eq. (57)] and truncated GIE [Eq. (53)], j th spectrum	$\mathbb{N}_1^{(j)}, \mathbb{N}_{1,P}^{(j)}, \mathbb{N}_{1,T}^{(j)}$
Time [Eq. (7)]	$\tau \equiv \omega t = De(t/\lambda_1)$
Weissenberg number [Eq. (5)]	$Wi \equiv \lambda_1 \dot{\gamma}^0$
Bessel function of first kind, m th order, argument Wi/De [Eq. (28)]	$J_m \equiv J_m(Wi/De)$
Spriggs exponent [Eqs. (42) and (43)]	α
Numbers of rows and columns in Ewoldt Grids [Eq. (31)]	r, c

Figure 1: Limiting behavior of our main result, Eq. (29) [with Eqs. (18)-(20)]. **Black** loop is when $\lambda_1 \omega \rightarrow 0$, and **red** curve is well-known expression for steady shear behavior (Eq. (67) of [38]).

Figure 2: Counterclockwise loops of *minus* dimensionless first normal stress differences, $-\mathbb{N}_1$, versus $W \cos 2\tau$, calculated for λ_2/λ_1 (a) 0, (b) $1/27$, (c) $1/9$ and (d) $1/3$ using $(De, Wi) = (1, 2)$. **Green** loops are [3,4] Padé approximant of Eq. (17) [Eq. (29) with Eqs. (18)-(20)], **red** are GIE [Eq. (17)], and **black** are exact solution [Eq. (21) with Eqs. (22)-(27)]. Padé approximant is more accurate than GIE from whence it came.

Figure 3: Counterclockwise loops of *minus* dimensionless first normal stress differences, $-\mathbb{N}_1$, versus $Wicos2\tau$, calculated for the special case of the corotational Maxwell fluid, Eq. (13) with $\lambda_2 = 0$, for $Wi/De = 1/10, 1/2, 3/4, 1, 5/4$ for each De . **Green** loops are [3,4] Padé approximant of Eq. (17) [Eq. (29) with Eqs. (18)-(20)], **red** are GIE [Eq. (17)], and **black** are exact solution [Eq. (21) with Eqs. (22)-(27)]. Padé approximant is more accurate than GIE from whence it came.

Figure 4: Counterclockwise loops of minus dimensionless first normal stress differences, $-\mathbb{N}_1$, versus $Wicos2\tau$, calculated for $\lambda_2/\lambda_1 = 1/27$, and for $Wi/De = 1/10, 1/2, 3/4, 1, 5/4$ for each De . **Green** loops are [3,4] Padé approximant of Eq. (17) [Eq. (29) with Eqs. (18)-(20)], **red** are GIE [Eq. (17)], and **black** are exact solution [Eq. (21) with Eqs. (22)-(27)]. Padé approximant is more accurate than GIE from whence it came.

Figure 5: Counterclockwise loops of *minus* dimensionless first normal stress differences, $-\mathbb{N}_1$, versus $W \cos 2\tau$, calculated for $\lambda_2/\lambda_1 = 1/9$, and for $Wi/De = 1/10, 1/2, 3/4, 1, 5/4$ for each De . **Green** loops are [3,4] Padé approximant of Eq. (17) [Eq. (29) with Eqs. (18)-(20)], **red** are GIE [Eq. (17)], and **black** are exact solution [Eq. (21) with Eqs. (22)-(27)]. Padé approximant is more accurate than GIE from whence it came.

Figure 6: Counterclockwise loops of *minus* dimensionless first normal stress differences, $-\mathbb{N}_1$, versus $W \cos 2\tau$, calculated for $\lambda_2/\lambda_1 = 1/3$, and for $Wi/De = 1/10, 1/2, 3/4, 1, 5/4$ for each De . **Green** loops are [3,4] Padé approximant of Eq. (17) [Eq. (29) with Eqs. (18)-(20)], **red** are GIE [Eq. (17)], and **black** are exact solution [Eq. (21) with Eqs. (22)-(27)]. Padé approximant is more accurate than GIE from whence it came.

Figure 7: 10×6 Ewoldt grids of [3,4] Padé approximant [Eq. (29) with Eqs. (18)-(20), **green**] compared with GIE [Eq. (17) with Eqs. (18)-(20) (**red**)], and with exact solution [Eq. (21) with Eqs. (22)-(27), **black**] with $\lambda_2/\lambda_1 = 0$, the special case of corotational Maxwell fluid. Disappearance of **red** loops indicates $\epsilon_T \gg 1$, and of **green**, singularities.

Figure 8: 10×6 Ewoldt grids of [3,4] Padé approximant [Eq. (29) with Eqs. (18)-(20), **green**] compared with GIE [Eq. (17) with Eqs. (18)-(20) (**red**)], and with exact solution [Eq. (21) with Eqs. (22)-(27), **black**] with $\lambda_2/\lambda_1 = 1/27$. Disappearance of **red** loops indicates $\epsilon_T \gg 1$, and of **green**, singularities.

Figure 9: 10×6 Ewoldt grids of [3,4] Padé approximant [Eq. (29) with Eqs. (18)-(20), **green**] compared with GIE [Eq. (17) with Eqs. (18)-(20) (**red**)], and with exact solution [Eq. (21) with Eqs. (22)-(27), **black**] with $\lambda_2/\lambda_1 = 1/9$. Disappearance of **red** loops indicates $\epsilon_T \gg 1$, and of **green**, singularities.

Figure 10: 10×6 Ewoldt grids of [3,4] Padé approximant [Eq. (29) with Eqs. (18)-(20), **green**] compared with GIE [Eq. (17) with Eqs. (18)-(20) (**red**)], and with exact solution [Eq. (21) with Eqs. (22)-(27), **black**] with $\lambda_2/\lambda_1 = 1/3$. Disappearance of **red** loops indicates $\epsilon_T \gg 1$, and of **green**, singularities.

Figure 11: Isopleths of $100\epsilon_p$ [Eq. (38)] subject to Eq. (39) for $\lambda_2/\lambda_1 = 0, 1/27, 1/9$ and $1/3$.

Figure 12: Isopleths of $100\epsilon_{\text{T}}$ [Eq. (37) subject to Eq. (40) for $\lambda_2/\lambda_1 = 0, 1/27, 1/9$ and $1/3$.

Figure 13: $\eta'(\omega)/\eta_0$ [Eq. (44) with Eqs. (41) and (42), **blue** curve] and $\eta''(\omega)/\eta_0$ [Eq. (45) with Eqs. (41) and (42), **magenta** curve] for generalized corotational Jeffreys fluid with Spriggs relations *versus* SAOS measurements (open circles) of TR480 HDPE at 170°C. The best-fit fluid parameters are $\eta_0 = 0.180 \text{ MPa} \cdot \text{s}$, $\lambda_1 = 60.0 \text{ s}$, $\lambda_2 = 0.500 \text{ s}$ and $\alpha = 2.31$.

Figure 14: Counterclockwise loops of *minus* dimensionless normal stress differences *versus* $W \cos 2\tau$. **Blue** open circles are LAOS measurements on TR480 HDPE melt at 170°C (see Figure 112. of [92]); $(\omega, \dot{\gamma}^0) = (31.4 \text{ rad/s}, 62.8 \text{ s}^{-1})$ or $(De, Wi) = (1880, 3770)$. **Red** loop is generalized GIE [Eq. (53) with Eqs. (54)-(56)], **green** loop is the generalized Padé approximant of Eq. (53) [Eq. (57) with Eqs. (54)-(56)], **black** loop is the generalized corotational Jeffreys fluid [Eqs. (46) with Eqs. (47)-(52)]. The best-fit fluid parameters are $\eta_0 = 0.180 \text{ MPa} \cdot \text{s}$, $\lambda_1 = 60.0 \text{ s}$, $\lambda_2 = 0.500 \text{ s}$ and $\alpha = 2.31$. Good agreement for $\Psi_1^d(\omega, \dot{\gamma}^0)$ and $\Psi'_{1,2}(\omega, \dot{\gamma}^0)$, but not for $\Psi''_{1,2}(\omega, \dot{\gamma}^0)$.

Figure 15: $\eta'(\omega)/\eta_0$ [Eq. (44) with Eqs. (41) and (42), **blue** curve] and $\eta''(\omega)/\eta_0$ [Eq. (45) with Eqs. (41) and (42), **magenta** curve] for generalized corotational Jeffreys fluid using Spriggs relation *versus* SAOS measurements (open circles) of 10% by weight solution of PIB dissolved in isobutylene oligomer at 20.4°C. The best-fit fluid parameters are $\eta_0 = 30.0 \text{ Pa} \cdot \text{s}$, $\lambda_1 = 0.470 \text{ s}$, $\lambda_2 = 5.00 \text{ ms}$ and $\alpha = 1.40$.

Figure 16: Counterclockwise loops of *minus* dimensionless normal stress differences *versus* $W \cos 2\tau$. **Blue** open circles are LAOS measurements on 10% by weight solution of PIB dissolved in isobutylene oligomer at 20.4°C; $(\omega, \dot{\gamma}^0) = (6.28 \text{ rad/s}, 9.87 \text{ s}^{-1})$ or $(De, Wi) = (2.95, 4.64)$. **Red** loop is generalized GIE [Eq. (53) with Eqs. (54)-(56)], **green** loop is the generalized Padé approximant of Eq. (53) [Eq. (57) with Eqs. (54)-(56)], **black** loop is the generalized corotational Jeffreys fluid [Eqs. (46) with Eqs. (47)-(52)]. The best-fit fluid parameters are $\eta_0 = 30.0 \text{ Pa} \cdot \text{s}$, $\lambda_1 = 0.470 \text{ s}$, $\lambda_2 = 5.00 \text{ ms}$ and $\alpha = 1.40$. Good agreement for $\Psi'_{1,2}(\omega, \dot{\gamma}^0)$ and $\Psi''_{1,2}(\omega, \dot{\gamma}^0)$, with $\Psi_1^d(\omega, \dot{\gamma}^0)$ slightly overpredicted.

VIII. REFERENCES

We alert readers to errata to References [2,27,35,41,43,53,70,97,98,100,101] given in Table I of [71] and Table III of [34]. Further, concerning [23], in Eq. (4) of [23], $\frac{9}{5}\lambda$ should be $\frac{9}{5}\lambda^3$.

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