Simple Accurate Expressions for Shear Stress in Large-Amplitude Oscillatory Shear Flow

Chaimongkol SAENGOW^{*,***}, Alan Jeffrey GIACOMIN^{*,**†}, Nidal KHALAF^{*,****} and Martin GUAY^{*}

^{*}Chemical Engineering Department

^{**}Mechanical and Materials Engineering Department Queen's University Kingston, Ontario, CANADA K7L 3N6

^{***}College of Integrated Science and Technology Rajamangala University of Technology Lanna Doisaket, Chiangmai, THAILAND 50220

^{****}Chemical Engineering Department American University of Beirut, Beirut, LEBANON 1107 2020

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Analytical solutions for shear stress in large-amplitude oscillatory shear flow (LAOS), for continuum or molecular models, often take the form of the first few terms of a power series in the shear rate amplitude. For corotational models, we get this truncated series using the Goddard-Miller integral expansion (GIE). Our previous work shows that the best Padé approximants for this truncated series, and specifically for the corotational Maxwell model in LAOS, can agree closely with the corresponding exact solution. We observe this close agreement, even for the Padé approximant for the series truncated after the fifth shear stress harmonic. In this paper, we begin with the extension of the GIE truncated after the next, seventh, order in the shear rate amplitude [*Phys. Fluids* 29, 043101 (2017)], and we then explore its Padé approximants. We uncover its best approximant, the [2,4], and compare it with both the GIE and the exact solution [*Macromol. Theory Simul.* 24, 352 (2015)]. We use Ewoldt grids to show the stunning accuracy of the [2,4] approximant in LAOS. We quantify this accuracy with an objective function and then map this function onto Pipkin space. We find the [2,4] approximant (from the GIE truncated after the seventh order in the shear rate amplitude) to be a simple accurate expression for the shear stress in LAOS. Our worked examples illustrate how researchers can use our new approximant reliably. For this, we use the Spriggs relations to extend the Padé approximant to multimode.

Key Words: Large-amplitude oscillatory shear (LAOS) / Corotational maxwell fluid / Padé approximants / Spriggs relations / Objective function / Pipkin space / Goddard-Miller integral expansion

1. INTRODUCTION

This paper addresses an emerging trend in the physics of fluids, the use of large-amplitude oscillatory shear flow for probing fluid elasticity (see [1]; Ch. 11 of [2]). More and more, physicists deepen their understanding of complex fluids this way. The present paper aims to help this growing community of scholars, both scientists and engineers, to exploit contemporary rheological theory that, with few exceptions³⁻⁵⁾, takes the form of expansions truncated after one of the first few harmonics of the shear stress, or of the normal stress difference responses⁶⁾.

Since its conception in 1935⁷⁻⁹⁾, 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 and the flow field in Fig. 1 of [3]):

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

and hence, to a corresponding cosinusoidal shear rate:

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

where, in this paper, symbols are defined in Table I and Table II (which follow Tables II and III of [10] closely, with adaptations for [11]). When sliding plate flow is used for large-amplitude oscillatory shear flow (LAOS), to avoid free boundary error, we require local shear stress transduction¹²⁻¹⁵⁾. In fact, the flow field in Eq. (1) is more commonly approximated with cone-plate flow where, in spherical coordinates (see Fig. A.8-2 of [58]):

* Corresponding author. Tel: +1-613-533-2768;
E-mail : giacomini@queensu.ca

$$\mathbf{v} = [v_r, v_\theta, v_\phi] \approx \left[0, 0, r \left[\frac{\pi - \theta}{2\theta_0} \right] \Omega_0 \cos \omega t \right] \quad (3)$$

which yields a flow field where $\dot{\gamma}_{\theta\phi}$ is nearly constant throughout the gap. Performing LAOS using cone-plate flow¹⁶⁾ requires local torque transduction over a region circumscribing the cone apex¹⁷⁻²¹⁾.

Using the characteristic relaxation time of the viscoelastic fluid, λ , we can nondimensionalize Eq. (2) (Eq. (2) of [10]):

$$\lambda \dot{\gamma}_{yx} = \lambda \dot{\gamma}^0 \cos \lambda \omega (t / \lambda) \equiv \text{WicosDe}(t / \lambda) \quad (4)$$

where the dimensionless Deborah and Weissenberg numbers are given by:

$$\text{De} \equiv \lambda \omega, \quad \text{Wi} \equiv \lambda \dot{\gamma}^0 \quad (5),(6)$$

each of which reflects both a property of the fluid, λ and property of the flow. In this paper, we follow Dealy *et al.*²²⁾ in plotting loops of shear stress *versus* shear rate, since these best bring out material nonlinearities as distortions from ellipticity²²⁻²⁴⁾.

Increasing either the Weissenberg number or the Deborah number in Eq. (4) causes the fluid response to further depart from Newtonian behavior (see Pipkin diagram, Fig. 2. of [4]).

When higher harmonics are observed in the shear stress response (see Eq. (21) of [25]):

$$\frac{\tau_{yx}(\omega t, \dot{\gamma}^0)}{\dot{\gamma}^0} = - \sum_{n=1, \text{odd}}^{\infty} \eta'_n(\omega, \dot{\gamma}^0) \cos n\omega t + \eta''_n(\omega, \dot{\gamma}^0) \sin n\omega t \quad (7)$$

we call the oscillatory experiment *large-amplitude*. Many important notations, alternative to Eqs. (7) have been introduced for analyzing the higher harmonics in the shear stress (see Eq. (3) of [26]); §9 and §10 of [10]).

The truncation of the power series of Eq. (7) can be usefully approximated with ratios of polynomials⁶⁾, and this is especially useful for molecular theory where extending these series can be prohibitively laborious (compare Eq. (20) of [25] with Eq. (82) of [27]; [28],[29]). We attribute the method for getting this set of polynomial ratios to Padé³⁰⁾ (see Chap. 1 of [31]). Elements of this set are not equally useful, and indeed, some or even all elements will sometimes be of little use. For instance, the limiting behaviors of the Padé approximants may be unphysical, or the roots of the denominators may introduce unphysical singularities. We call each ratio the

Table I. Dimensional variables

Name	Unit	Variable
Angular velocity [Eq. (2)]	t^{-1}	ω
Angular velocity, amplitude, cone attachment	t^{-1}	Ω_0
Cartesian coordinate	L	x, y, z
Spherical coordinates	L	r
Extra stress, yx component*	M/Lt^2	τ_{yx}
Extra stress, yx component*, n th measured harmonic [Eq. (10)]	M/Lt^2	σ_n^m
Extra stress, yx component*, n th predicted harmonic [Eq. (11)]	M/Lt^2	σ_n^p
Extra stress, yx component*, j th spectrum [Eq. (36)]	M/Lt^2	τ_j
Extra stress tensor [Eq. (13)]	M/Lt^2	$\boldsymbol{\tau}$
Rate of strain tensor [Eq. (15)]	t^{-1}	$\dot{\gamma}$
Relaxation time, j th spectrum [Eq. (34)]	t	λ_j
Relaxation time, the longest spectrum [Eq. (4)]	t	$\lambda \equiv \lambda_1$
Shear rate, yx component [Eq. (2)]	t^{-1}	$\dot{\gamma}_{yx}$
Shear rate, $\theta\phi$ component	t^{-1}	$\dot{\gamma}_{\theta\phi}$
Shear rate, amplitude [Eq. (4)]	t^{-1}	$\dot{\gamma}^0$
Loss and storage viscosities, n th harmonic [Eq. (7)]	M/Lt	η'_n, η''_n
Time	t	t
Velocity vector [Eq. (1)]	L/t	$\mathbf{v}$
Velocity, i th component [Eq. (1)]	L/t	v_i
Viscosity, j th spectrum [Eq. (34)]	M/Lt	η_j
Viscosity, zero shear [Eq. (13)]	M/Lt	η_0
Vorticity tensor [Eq. (16)]	t^{-1}	$\boldsymbol{\omega}$

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

*Where τ_j 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 [58], or pp. 24–25 of [59]).

[x,y] Padé approximant, where x is the order of the numerator, and y, of the denominator.

For polymeric liquids, these higher harmonics are commonly observed when (see Eq. (11) [32] and Fig. 14 of [32]):

$$[\text{Wi} > 1] \cap \left[\frac{\text{Wi}}{\text{De}} > 1 \right] \quad (8)$$

Eq. (8) is thus our working definition of LAOS, and which improves upon previous definitions ([3,33,34]; Eq. (9) of [4]; Eq. (8) of [10]). With recent advances¹⁶⁾, rheometers for experiments satisfying Eq. (8) are now available. Conducting

Table II. Dimensionless variables and groups

Name	Symbol
Cone angle [Eq. (3)]	θ_0
Deborah number [Eq. (5)]	De
Degree of Padé approximant, [numerator,denominator]	[x,y]
Error (proposed by Jeyaseelan and Giacomini) [Eq. (9)]	Γ
Error [Eq. (12)]	ϵ
Error, Goddard integral expansion approximation [Eq. (30)]	ϵ_T
Error, Padé approximant [Eq. (31)]	ϵ_P
Functions in Eqs. (27) and (28)	I_1, I_2
Functions in Eqs. (20)–(23)	A, B, C, D
Relaxation time spectrum constant [Eq. (34)]	α
Shear stress response, any approximate solution	$\tilde{\mathbb{S}}$
Shear stress response, exact solution [Eq. (26)]	$\mathbb{S}$
Shear stress response, exact solution, absolute maximum value	$\mathbb{S}_{\max}$
Shear stress response, Padé approximant [Eq. (24)]	$\mathbb{S}_P$
Shear stress response, truncated Goddard integral expansion [Eq. (19)]	$\mathbb{S}_T$
Spherical coordinates, latitudinal and longitudinal	θ, φ
Weissenberg number [Eq. (5)]	Wi

experiments satisfying Eq. (8) is now commonplace for exploring the nonlinear physics of polymeric liquids. The intersection Eq. (8) is consistent with the graphical definitions of large-amplitude of Fig. 5-27 of [35], Fig. 1. of [36] or the figure in [37].

In the study of complex fluids, predicting and anticipating fluid behaviors is a must. For fluids, subjected to LAOS, Giacomini arrived at the first approximate solutions for the corotational Maxwell fluid from first principles using a truncation of the Goddard-Miller integral expansion (GIE). This gave rheologists a first glimpse of the nonlinear behaviors of complex fluids, but the region of accuracy for this truncation was not given. Other forms of approximate solutions including a simple scalar model³⁸⁾ and Padé approximants⁶⁾ were also explored. Exact solutions, first for a single corotational model³⁾, but more recently, for the entire corotational framework, were derived recently^{4, 5)}. Whereas the GIE truncation accuracy decreases with increasing Wi, exact solutions can be used to explore any conditions of LAOS, $(\omega, \dot{\gamma}^0)$.

Previously, for assessing the accuracy of an LAOS measurement with a theoretical prediction, Jeyaseelan and Giacomini proposed (Eq. (21) of [39]):

$$\Gamma \equiv \sum_{n, \text{odd}} \frac{1}{n} \left(\frac{\sigma_n^m - \sigma_n^p}{\sigma_n^m} \right)^2 \quad (9)$$

where σ_n^m is the amplitude of the n th measured shear stress harmonic, and σ_n^p , predicted:

$$\sigma_n^m \equiv \dot{\gamma}^0 \sqrt{(\eta_n^m)^2 + (\eta_n^{\prime m})^2}, \quad \sigma_n^p \equiv \dot{\gamma}^0 \sqrt{(\eta_n^p)^2 + (\eta_n^{\prime p})^2} \quad (10), (11)$$

The objective function Γ is for comparing one Fourier series with another. In this paper, we compare Padé approximants with an exact solution for the shear stress, neither of which are Fourier series. We thus create the following new *objective function*:

$$\epsilon \equiv \sqrt{\oint (\tilde{\mathbb{S}} - \mathbb{S})^2 / \mathbb{S}_{\max}^2 d\tau} \quad (12)$$

where $\tilde{\mathbb{S}}$ is an approximation to the shear stress, and $\mathbb{S}$, an exact solution. Whereas Eq. (9) [with Eqs. (10) and (11)] includes only the amplitudes of the shear stress harmonics, Eq. (12) includes every detail of the shear stress wave form. We are thus proposing Eqs. (30) and (31) as a significant improvement over Eq. (9). We will apply Eq. (12) twice in Subsection 2.3.

2. ANALYSIS

In this section, we assess the accuracy of the truncated GIE (Eq. (82) of [4]) to the corotational Maxwell model in LAOS, and of its corresponding [2,4] Padé approximant. We choose corotational Maxwell because this two-parameter model (η_0 and λ) is the simplest constitutive model that is relevant to LAOS. By *relevant*, we mean that the model predicts higher harmonics for the shear stress, and for the normal stress differences. Further, when multiple relaxation times are used, corotational Maxwell has previously been found to be accurate for the measured shear stress response of molten plastics (see §11 of [10]; Section 7.1.1 of [3]; Section 5.2 of [40]; [32,41]).

2.1 Corotational maxwell model

The corotational framework has been studied extensively, both in theory [3,4,5,10] and in its applications [32,42,43,44,45]. The simplest special case of this framework is the corotational Maxwell fluid:

$$\tau + \lambda \frac{\mathcal{D}\tau}{\mathcal{D}t} = -\eta_0 \dot{\gamma} \quad (13)$$

where λ is the characteristic time of the fluid, and η_0 is its zero

shear viscosity. In Eq. (13) the corotational derivative of the extra stress tensor is:

$$\frac{\mathcal{D}\boldsymbol{\tau}}{\mathcal{D}t} \equiv \frac{D\boldsymbol{\tau}}{Dt} + \frac{1}{2}\{\boldsymbol{\omega} \cdot \boldsymbol{\tau} - \boldsymbol{\tau} \cdot \boldsymbol{\omega}\} \quad (14)$$

where $D\boldsymbol{\tau}/Dt$ is the substantial derivative and:

$$\dot{\boldsymbol{\gamma}} \equiv \nabla \mathbf{v} + (\nabla \mathbf{v})^\dagger, \quad \boldsymbol{\omega} \equiv \nabla \mathbf{v} - (\nabla \mathbf{v})^\dagger \quad (15), (16)$$

are the rate-of-strain, and vorticity tensors. For an extensive discussion of corotational models and their applications, see Chapters 7 and 8 of [46], and also [47,48,49,50,51].

For any simple shear flow (see Eq. (19) of [52]), the rightmost term in Eq. (14) is given by:

$$\frac{1}{2}\{\boldsymbol{\omega} \cdot \boldsymbol{\tau} - \boldsymbol{\tau} \cdot \boldsymbol{\omega}\} = \frac{1}{2} \begin{bmatrix} -2\tau_{yx} & \tau_{xx} - \tau_{yy} & 0 \\ \tau_{xx} - \tau_{yy} & 2\tau_{yx} & 0 \\ 0 & 0 & 0 \end{bmatrix} \dot{\gamma}_{yx}(t) \quad (17)$$

and with the use of Eq. (1), we can nondimensionalize Eq. (15) as:

$$\lambda \dot{\boldsymbol{\gamma}} = \begin{bmatrix} 0 & \lambda \dot{\gamma}^0 \cos \lambda \omega(t/\lambda) & 0 \\ \lambda \dot{\gamma}^0 \cos \lambda \omega(t/\lambda) & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad (18)$$

$$\equiv \begin{bmatrix} 0 & W \cos De(t/\lambda) & 0 \\ W \cos De(t/\lambda) & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}$$

where De and Wi are the Deborah and Weissenberg numbers. Letting $\tau \equiv De(t/\lambda)$, the truncated GIE to Eq. (13) for the shear stress in LAOS has been given by (Eq. (82) of [4]):

$$\mathbb{S}_T \equiv \frac{\tau_{yx}}{\eta_0 \dot{\gamma}^0} = A + B Wi^2 + C Wi^4 + D Wi^6 + \dots \quad (19)$$

where:

$$A \equiv -\left(\frac{\cos \tau + De \sin \tau}{1 + De^2} \right) \quad (20)$$

$$B \equiv \frac{1}{4} \left[\frac{3 \cos \tau + 6 De \sin \tau}{(1 + De^2)(1 + 4 De^2)} + \frac{(1 - 11 De^2) \cos 3\tau + 6 De(1 - De^2) \sin 3\tau}{(1 + De^2)(1 + 4 De^2)(1 + 9 De^2)} \right] \quad (21)$$

$$C \equiv \frac{-1}{8} \left[\frac{5 \cos \tau + 15 De \sin \tau}{(1 + De^2)(1 + 4 De^2)(1 + 9 De^2)} + \frac{(5 - 130 De^2) \cos 3\tau + (45 - 120 De^2) De \sin 3\tau}{2(1 + De^2)(1 + 4 De^2)(1 + 9 De^2)(1 + 16 De^2)} + \frac{(1 - 85 De^2 + 274 De^4) \cos 5\tau + (15 - 225 De^2 + 120 De^4) De \sin 5\tau}{2(1 + De^2)(1 + 4 De^2)(1 + 9 De^2)(1 + 16 De^2)(1 + 25 De^2)} \right] \quad (22)$$

$$D \equiv \frac{1}{64} \left[\frac{35 \cos \tau + 140 De \sin \tau}{(1 + De^2)(1 + 4 De^2)(1 + 9 De^2)(1 + 16 De^2)} + \frac{21(1 - 47 De^2) \cos 3\tau + 252(1 - 5 De^2) De \sin 3\tau}{(1 + De^2)(1 + 4 De^2)(1 + 9 De^2)(1 + 16 De^2)(1 + 25 De^2)} + \frac{7(1 - 155 De^2 + 1044 De^4) \cos 5\tau + 140(1 - 29 De^2 + 36 De^4) De \sin 5\tau}{(1 + De^2)(1 + 4 De^2)(1 + 9 De^2)(1 + 16 De^2)(1 + 25 De^2)(1 + 36 De^2)} + \frac{(1 - 322 De^2 + 6769 De^4 - 13068 De^6) \cos 7\tau + 28(1 - 70 De^2 + 469 De^4 - 180 De^6) De \sin 7\tau}{(1 + De^2)(1 + 4 De^2)(1 + 9 De^2)(1 + 16 De^2)(1 + 25 De^2)(1 + 36 De^2)(1 + 49 De^2)} \right] \quad (23)$$

Eq. (19) improved upon Eq. (58) of [10,53] by one order of Wi . In this paper, we find the best Padé approximant by exploring the upper left corner of the Padé table (see after Eq. (31) in [6]). We arrive at the best Padé approximant of Eq. (19) as the [2,4] Padé approximant. By the *best* [x,y], we mean the approximant, over the widest area of the Pipkin space, for which no singularity is observed over the range $0 \leq \omega t < 2\pi$, giving dimensionless shear stress *versus* shear rate loops having two-fold symmetry, and having the lowest value of $(x+y)$. This best Padé approximant is the [2,4]:

$$\mathbb{S}_P(\tau) \equiv \frac{\tau_{yx}}{\eta_0 \dot{\gamma}^0} = \frac{A^2 C - AB^2 + (-A^2 D + 2ABC - B^3) Wi^2}{AC - B^2 + (-AD + BC) Wi^2 + (BD - C^2) Wi^4} \quad (24)$$

where A, B, C and D are defined in Eqs. (20)–(23). Eq. (24) [with Eqs. (20)–(23)] is the main result of this paper. This main result was arrived at for truncations after Wi^6 , which includes harmonics up to the seventh. It goes without saying that our main result will be equally useful for lower order truncations. For instance, Eq. (20) of [25] offers a Wi^4 truncation for a rigid dumbbell suspension ($D = 0$), or Eq. (2.17) of [29] offers a Wi^2 truncation for a Kramers chain ($C = D = 0$). In our previous paper, we reported on a best approximant (Eq. (35) of [6]) to the GIE for the corotational Maxwell fluid truncated after Wi^4 . If we set $D = 0$, our new [2,4] approximant to the GIE truncated after Wi^6 , Eq. (24)[with Eqs. (20)–(23)], does not (and need not) reduce to Eq. (35) of [6].

All Padé approximants will be singular when their denominators go to zero, and one must be mindful of this when using them. If denominator singularities in Eq. (24) arise over the range $0 \leq \omega t < 2\pi$, these will be at most four roots of:

$$AC - B^2 + (-AD + BC) Wi^2 + (BD - C^2) Wi^4 = 0 \quad (25)$$

Figure 1 shows that the limiting behaviors of our main result, Eq. (24) [with Eqs. (20)–(23)] are consistent with the well-known expressions for the corotational Maxwell model for small-amplitude oscillatory shear flow (SAOS), Eq. (59) of [10], and for steady shear flow (Eq. (76) of [4]).

The exact analytical solution to Eq. (13) for the shear stress is (Eq. (65) [with Eqs. (66) and (67)] of [4] where

Fig. 1 Limiting behaviors of our main result, Eq. (24) [with Eqs.(20)–(23)], green loop is when $\lambda\dot{\gamma}^0 \rightarrow 0$, and green curve, $\lambda\omega \rightarrow 0$. Red loop is well-known linear viscoelastic expression (Eq. (59) of [10]) and red curve, steady shear (Eq. (76) of [4]).

$$\lambda_2 = \mu_0 = \mu_1 = \mu_2 = \nu_1 = \nu_2 = 0):$$

$$\mathbb{S}(\tau) \equiv \frac{\tau_{yx}}{\eta_0 \dot{\gamma}^0} = -\frac{1}{De} e^{\frac{\tau}{De}} \sin\left(\frac{Wi}{De} \sin \tau\right) I_1 - \frac{1}{De} e^{\frac{\tau}{De}} \cos\left(\frac{Wi}{De} \sin \tau\right) I_2 \quad (26)$$

where:

$$I_1 = \frac{4De^2}{Wi} e^{\frac{\tau}{De}} \sum_{k=1}^{\infty} \left[\frac{k J_{2k}}{1 + 4De^2 k^2} (-2De k \cos 2k\tau + \sin 2k\tau) \right] \quad (27)$$

$$I_2 = \frac{2De^2}{Wi} e^{\frac{\tau}{De}} \sum_{k=1}^{\infty} \left[\frac{(2k-1) J_{2k-1}}{1 + (2k-1)^2 De^2} (\cos[2k-1]\tau + De(2k-1) \sin[2k-1]\tau) \right] \quad (28)$$

where $J_k \equiv J_k(Wi/De)$ is the Bessel function of the k th order of first kind. This exact solution will serve as the standard in this paper wherever accuracy is discussed. We will use Eqs. (19) and (24), with Eq. (26) [with Eqs. (27) and (28)] to construct the Ewoldt grids in Subsection 2.2, and to assess accuracy throughout Subsection 2.3.

2.2 Ewoldt grids

In this paper, we illustrate the material response in LAOS with dimensionless loops of the shear stress *versus* shear rate, each of which is constructed for specific conditions (De, Wi). We can then miniaturize these loops and map them in rows and columns onto Pipkin space, that is, onto a map of Wi *versus* De (see Fig. 2. of [4]). We call this grid of loops an *Ewoldt grid* (see Fig. 4. and Fig. 8. of [54]; Figs. 4-4 through 4-6 in [55]; Fig. 1. in [56]).

The general expression for each cell in the grids is given

Fig. 2 10×6 Ewoldt grids of [2,4] Padé approximant [Eq. (24) with Eqs. (20)–(23), green] *versus* Eq. (19) [with Eqs. (20)–(23)], red, and also *versus* exact solution [Eq. (26) with Eqs. (27) and (28), black]. Counterclockwise loops of *minus* dimensionless shear stress, $-\mathbb{S}$, *versus* dimensionless shear rate, $\lambda\dot{\gamma}$, calculated for the corotational Maxwell fluid. Disappearance of red loops indicates $\epsilon_T \gg 1$, and of green, singularities.

by:

$$\left[\frac{\mathbb{S}(De, Wi)}{\mathbb{S}_{\max}} \right]_{i,j} = \frac{\mathbb{S}(De + 2(i-1), Wi)}{\mathbb{S}_{\max}} + 2(j-1) \quad (29)$$

where $i = 1$ and $j = 1$ is the lowest left cell of the grid, and where $\mathbb{S}_{\max}$ is the peak shear stress given by the exact solution, Eq. (26) [with Eqs. (27) and (28)], for each of the value of De and Wi in LAOS. We will thus use Eqs. (19), (24), and Eq. (26) [with Eqs. (27) and (28)] to plot Fig. 2.

2.3 Objective function

Here we apply Eq. (12) to (i) the truncated GIE (Eq. (19) [with Eqs. (20)–(23)]) to give:

$$\epsilon_T \equiv \sqrt{\oint \frac{(\mathbb{S}_T - \mathbb{S})^2}{\mathbb{S}_{\max}^2} d\tau} = \frac{1}{\mathbb{S}_{\max}} \sqrt{\int_0^{2\pi} (\mathbb{S}_T - \mathbb{S})^2 d\tau} \quad (30)$$

and (ii) the [2,4] Padé approximant (Eq. (24) [with Eqs. (20)–(23)]) to give:

$$\epsilon_P \equiv \sqrt{\oint \frac{(\mathbb{S}_P - \mathbb{S})^2}{\mathbb{S}_{\max}^2} d\tau} = \frac{1}{\mathbb{S}_{\max}} \sqrt{\int_0^{2\pi} (\mathbb{S}_P - \mathbb{S})^2 d\tau} \quad (31)$$

Fig. 3 Eq. (19) (red) *versus* its [2,4] Padé approximant [Eq. (24) with Eqs. (20)–(23), green], and also *versus* exact solution [Eq. (26) with Eqs. (27) and (28), black]. Counterclockwise loops of *minus* dimensionless shear stress, $-\mathbb{S}$, *versus* dimensionless shear rate, $\lambda\dot{\gamma}$, calculated for the corotational Maxwell fluid with $Wi/De = 4$ and $De = 1$. For Eq. (19), red, $\epsilon_T \gg 1$.

Fig. 4 [2,4] Padé approximant of Eq. (19) [Eq. (24) with Eqs. (20)–(23), green] *versus* Eq. (19), red, and also *versus* exact solution [Eq. (26) with Eqs. (27) and (28), black]. Counterclockwise loops of *minus* dimensionless shear stress, $-\mathbb{S}$, *versus* dimensionless shear rate, $\lambda\dot{\gamma}$, calculated for the corotational Maxwell fluid with $Wi/De = 2$ and $De = 2$. Padé approximant shows better accuracy than the Goddard integral approximation.

We thus quantify the inaccuracies of Items (i) and (ii) with the objective functions, ϵ_T and ϵ_p , and then map these onto Pipkin space. In Eqs. (30) or (31), $\mathbb{S}_T$ is given by Eq. (19) and $\mathbb{S}_p$, by Eq. (24), and $\mathbb{S}$, by Eq. (26) [with Eqs. (27) and (28)]. We thus next map the ϵ_T and ϵ_p error isopleths as contours in Pipkin space. We use Eq. (30) [with Eqs. (19) and (26)] to plot Fig. 6, and Eq. (31) [with Eqs. (24) and (26)], to plot Fig. 5.

3. DISCUSSION

From the Ewoldt grid that is Fig. 2, we learn that our main result, the [2,4] Padé approximant (Eq. (24) [with Eqs. (20)–(23)]), yields loop singularity when:

$$[Wi > 0.1] \cap \left[\frac{Wi}{De} > 2 \right] \quad (32)$$

Over this same region, from this same Ewoldt grid, we also observe $\epsilon_T \gg 1$ for the truncated GIE. Figure 3 illustrates one such example at $(De, Wi) = (1, 4)$ of $\epsilon_T \gg 1$. Our Ewoldt grid, Fig. 2, thus omits both Padé approximant predictions with loop singularities and truncated GIE loop predictions with $\epsilon_T \gg 1$. We find no examples where the Padé approximant is singular and where $\epsilon_T \gg 1$ does not obtain. We do, however, find examples where $\epsilon_T \gg 1$ and the Padé approximant is not singular (see Fig. 3). Otherwise put, we find that Padé ap-

proximant singularity and truncated GIE inaccuracy ($\epsilon_T \gg 1$) do not go hand in hand. This yields the useful implication that the Padé can be inaccurate when it is not singular. When using our new [2,4] approximant, we must thus rely heavily on the union Eq. (33).

Comparing ϵ_p with ϵ_T (Fig. 5 *versus* Fig. 6), we see that our main result, the [2,4] Padé approximant (Eq. (24) [with Eqs. (20)–(23)]), is more accurate than the truncated GIE from whence it came. For example, the contour $\epsilon_T = 0.2$ (see Fig. 6) is well below $\epsilon_p = 0.2$ (see Fig. 5).

Furthermore, we find our [2,4] Padé approximant to be useful when:

$$[Wi < 0.1] \cup \left[\frac{Wi}{De} \leq 2 \right] \quad (33)$$

By *useful* at (De, Wi) , we mean the approximant exhibits no singularity over the range $0 \leq \omega t < 2\pi$, and gives dimensionless shear stress *versus* shear rate loops having two-fold symmetry. To illustrate the stunning agreement between our main result and the exact solution, we plot Fig. 4 for just one cell, $(De, Wi) = (2, 4)$, from the Ewoldt grid that is Fig. 2.

From Fig. 2, Fig. 5 and Fig. 6, we learn that the Goddard integral expansion always departs from the exact solution before the Padé approximant does. From Fig. 2, Fig. 5 and Fig. 6, we also learn that increasing De always improves the accuracy of our main result, Eq. (24) [with Eqs. (20)–(23)].

4. APPLICATION: LOOP ANALYSIS

Here we generalize and apply our main result to the measured behavior of molten high-density polyethylene (HDPE) in LAOS at 160 °C (see experimental detailed in §11. of [10]), and specifically at $\omega = 0.725 \text{ rad/s}$, $\dot{\gamma}^0 = 0.906 \text{ s}^{-1}$. We begin by following §7a. of [10] and replace the 11 relaxation time discrete spectrum of Eqs. (113) and (114) (with Table IV of [10]) with the Spriggs relations (Eqs. (6.1-14) and (6.1-15) of [46]):

$$\eta_j = \eta_0 \lambda_j / \sum_{j=1}^{\infty} \lambda_j, \quad \lambda_j = \lambda / j^\alpha \quad (34), (35)$$

where $\eta_0 = 1.51 \times 10^5 \text{ Pa}\cdot\text{s}$ is the zero shear viscosity, and $\lambda \equiv \lambda_1 = 8.41 \text{ s}$ is the longest relaxation time, and $\alpha = 2.41$, and where infinite spectrum contribute nothing to the generalized shear stress expression. Since j th viscosity increases with relaxation time, the longest relaxation time contributes the most to the generalized shear stress expression. Eqs. (34) and (35) thus provide us with an infinity of relaxation times, and unequivocally, with a longest relaxation time, λ . We can thus use this λ to nondimensionalize experimental conditions, and thus for this particular application, $De = 6.10$ and $Wi = 7.62$. Since these values satisfy Eq. (33), we can expect to use the [2,4] Padé approximant for the shear stress without singularity.

We next generalized Eq. (24) for multimode to:

$$\frac{\tau_{yx}}{\dot{\gamma}^0} = \sum_{j=1}^{\infty} \frac{\tau_j}{\dot{\gamma}^0} = \sum_{j=1}^{\infty} \eta_j \frac{A^2 C - AB^2 + (-A^2 D + 2ABC - B^3)(\lambda_j \dot{\gamma}^0)^2}{AC - B^2 + (-AD + BC)(\lambda_j \dot{\gamma}^0)^2 + (BD - C^2)(\lambda_j \dot{\gamma}^0)^4} \quad (36)$$

where:

$$A \equiv - \left(\frac{\cos \tau + \lambda_j \omega \sin \tau}{1 + \lambda_j^2 \omega^2} \right) \quad (37)$$

$$B \equiv \frac{1}{4} \left[\frac{3 \cos \tau + 6 \lambda_j \omega \sin \tau}{(1 + \lambda_j^2 \omega^2)(1 + 4 \lambda_j^2 \omega^2)} + \frac{(1 - 11 \lambda_j^2 \omega^2) \cos 3\tau + 6(\lambda_j \omega)(1 - \lambda_j^2 \omega^2) \sin 3\tau}{(1 + \lambda_j^2 \omega^2)(1 + 4 \lambda_j^2 \omega^2)(1 + 9 \lambda_j^2 \omega^2)} \right] \quad (38)$$

$$C \equiv \frac{-1}{8} \left[\frac{5 \cos \tau + 15 \lambda_j \omega \sin \tau}{(1 + \lambda_j^2 \omega^2)(1 + 4 \lambda_j^2 \omega^2)(1 + 9 \lambda_j^2 \omega^2)} + \frac{(5 - 130 \lambda_j^2 \omega^2) \cos 3\tau + (45 - 120 \lambda_j^2 \omega^2) \lambda_j \omega \sin 3\tau}{2(1 + \lambda_j^2 \omega^2)(1 + 4 \lambda_j^2 \omega^2)(1 + 9 \lambda_j^2 \omega^2)(1 + 16 \lambda_j^2 \omega^2)} + \frac{(1 - 85 \lambda_j^2 \omega^2 + 274 \lambda_j^4 \omega^4) \cos 5\tau + (15 - 225 \lambda_j^2 \omega^2 + 120 \lambda_j^4 \omega^4) \lambda_j \omega \sin 5\tau}{2(1 + \lambda_j^2 \omega^2)(1 + 4 \lambda_j^2 \omega^2)(1 + 9 \lambda_j^2 \omega^2)(1 + 16 \lambda_j^2 \omega^2)(1 + 25 \lambda_j^2 \omega^2)} \right] \quad (39)$$

$$D \equiv \frac{1}{64} \left[\frac{35 \cos \tau + 140 \lambda_j \omega \sin \tau}{(1 + \lambda_j^2 \omega^2)(1 + 4 \lambda_j^2 \omega^2)(1 + 9 \lambda_j^2 \omega^2)(1 + 16 \lambda_j^2 \omega^2)} + \frac{21(1 - 47 \lambda_j^2 \omega^2) \cos 3\tau + 252(1 - 5 \lambda_j^2 \omega^2) \lambda_j \omega \sin 3\tau}{(1 + \lambda_j^2 \omega^2)(1 + 4 \lambda_j^2 \omega^2)(1 + 9 \lambda_j^2 \omega^2)(1 + 16 \lambda_j^2 \omega^2)(1 + 25 \lambda_j^2 \omega^2)} + \frac{7(1 - 155 \lambda_j^2 \omega^2 + 1044 \lambda_j^4 \omega^4) \cos 5\tau + 140(1 - 29 \lambda_j^2 \omega^2 + 36 \lambda_j^4 \omega^4) \lambda_j \omega \sin 5\tau}{(1 + \lambda_j^2 \omega^2)(1 + 4 \lambda_j^2 \omega^2)(1 + 9 \lambda_j^2 \omega^2)(1 + 16 \lambda_j^2 \omega^2)(1 + 25 \lambda_j^2 \omega^2)(1 + 36 \lambda_j^2 \omega^2)} + \frac{(1 - 322 \lambda_j^2 \omega^2 + 6769 \lambda_j^4 \omega^4 - 13068 \lambda_j^6 \omega^6) \cos 7\tau + 28(1 - 70 \lambda_j^2 \omega^2 + 469 \lambda_j^4 \omega^4 - 180 \lambda_j^6 \omega^6) \lambda_j \omega \sin 7\tau}{(1 + \lambda_j^2 \omega^2)(1 + 4 \lambda_j^2 \omega^2)(1 + 9 \lambda_j^2 \omega^2)(1 + 16 \lambda_j^2 \omega^2)(1 + 25 \lambda_j^2 \omega^2)(1 + 36 \lambda_j^2 \omega^2)(1 + 49 \lambda_j^2 \omega^2)} \right] \quad (40)$$

Eqs. (34) are called the Spriggs relations, and for more on these see §6.1 of [46], Appendix of [57], and §8.5 of [59]). Figure 7 shows that the [2,4] Padé approximant for the generalized corotational Maxwell model agrees closely with the measured behavior of the molten plastic in LAOS.

5. CONCLUSION

In this paper, we focus on the shear stress response of polymeric liquids subject to LAOS, and specifically, on expansions for this shear stress truncated after Wi^6 . These expansions include shear stress harmonics up to and including the seventh. We seek a simple way of improving the accuracies of these expansions by the method of Padé.

Fig. 5 Isopleths of $100\epsilon_p$ [Eq. (31)] subject to Eq. (33).

Fig. 6 Isopleths of $100\epsilon_r$ [Eq. (30)] subject to Eq. (33).

Fig. 7 LAOS measurements of HDPE melts at 160 °C (blue circles) versus the best fit of the generalized Padé approximant of the Goddard integral expansion approximation [Eq. (36) with Eqs. (37)–(40), green loop] for the generalized corotational Maxwell, using the Spriggs relations [Eqs. (36) with Eq. (34)] with $\eta_0 = 1.51 \times 10^5 \text{ Pa}\cdot\text{s}$, $\lambda = 8.41 \text{ s}$ and $\alpha = 2.41$. The outermost ellipse is linear viscoelasticity [$\lambda\dot{\gamma}^0 \rightarrow 0$ of Eqs. (36) with Eq. (34)], and the inner curve is steady shear flow behavior, $\lambda\omega \rightarrow 0$. The experimental conditions are $\omega = 0.725 \text{ rad/s}$ and $\dot{\gamma}^0 = 0.906 \text{ s}^{-1}$ [De = 6.10 and Wi = 7.62].

We discover a new Padé approximant of the Goddard integral expansion truncated after Wi^6 , and this [2,4] approximant is the main result of this paper Eq. (24) [with Eqs. (20)–(23)]. So long as its denominator is not singular, our [2,4] approximant is always more accurate than the Goddard integral expansion from whence it came. With considerable effort, molecular theory yields truncated expansions in Wi . We thus expect our [2,4] approximant to be especially useful to those applying the results of polymer molecular theory.

We find our [2,4] approximant to be singular wherever the Goddard integral expansion from whence it came becomes inaccurate. However, since [2,4] approximant singularity and its inaccuracy do not go hand in hand, as a practical matter, we provide the union of inequalities, Eq. (33), as a companion relation for our main result.

For those finding the infinities of Bessel functions in Eqs. (27) and (28) to be cumbersome, our [2,4] approximant offers the additional advantage of simplicity over the exact solution, Eq. (26) [with Eqs. (27) and (28)], so long as Eq. (33) holds. Section 4 helps researchers to use our main result. Moreover, in Section 4, we compare our [2,4] approximant with experimental data by first generalizing it to multimode using the Spriggs relations. We find that, when generalized, the Padé approximant agrees closely with published experimental measurements of high-density polyethylene melts.

In this paper, we derived the [2,4] Padé approximant from the GIE for the corotational Maxwell fluid for the shear stress in LAOS, truncated after Wi^6 (Eq. (19) [with Eqs. (20)–(23)]). Following the method of §8 of [10], the GIE for the corotational Jeffreys fluid for the shear stress in LAOS, truncated after Wi^6 (Eq. (83) of [4]), has been derived recently. Following the methods herein, one could thus easily explore the [2,4] Padé approximant for the GIE for the corotational Jeffreys fluid (Eq. (83) of [4]). This derivation, we shall leave for another day.

The same approach used in this paper could be applied to the normal stress differences, for example Eq. (74) of [5], for the corotational Maxwell fluid. This too, we shall leave for another day.

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APPENDIX: USEFUL ERRATA TO CITED ARTICLES

In this appendix, in Table III, we gather useful errata to our cited literature.

Table III. Errata to Cited Articles

Ref.	Errata
[3]	In Eqs. (47) and (48), each “ τ ” in the integrands should be “ τ' ”
[4]	In column 4 of rows 12 and 13, $\lambda_2 = \mu_0 = \mu_1$ should be $\mu_1 = -\lambda_1$; $\lambda_2 = \mu_0$ and $\mu_1 = \lambda_1$; $\lambda_2 = \mu_0$.
[6]	In Eq. (35), “ $S(\tau)$ ” should be “ $-S(\tau)$ ”
[8]	The footnote on p. 1583, “ ¹⁵ A. Gemant, <i>Naturwiss.</i> , 1935, 23, 406.” should be “ ¹⁵ A. Gemant, <i>Naturwiss.</i> , 1935, 25, 406.”

[10]	After Eq. (20), Ref. [10] should be [13]; Eq. (65) should be $We/De > \sqrt[n]{(n+1)!}$; after Eq. (65), $\sqrt[3]{3!}$ should be $\sqrt[4]{5!}$, and $\gamma_0 > 1.82$ should be $\gamma_0 > 3.31$; in Eq. (66), “20De ² ” and “10De ² –50De ⁴ ” should be “20De” and “(10De–50De ³) De” and so Fig. 15 through Fig. 17 of [60] replace Figs. 5–7; on the ordinates of Figs. 5–7, $\frac{1}{2}$ should be 2; in Eqs. (76) and (77), Ψ' and Ψ'' should be Ψ'_1 and Ψ''_1 ; throughout, Ψ'_1 , Ψ''_1 and Ψ'''_1 should be Ψ'_1 , Ψ''_1 and Ψ'''_1 ; after Eq. (119), “($\zeta\alpha$)” should be “ $\zeta(\alpha)$ ”; in Eq. (127), “cos τ ” should be “(–cos τ)”; after Eq. (136), “Eq. (134)” should be “Eqs. (133) and (135)” and “Eq. (135)” should be “Eqs. (134) and (135)”; after Eq. (143), $ \tau_{yx} $ should be $\dot{\gamma}$; in Eq. (147), “n–1” should be “n = 1”; in Eqs. (181) and (182), “1,21” should be “1,2”; after Eq. (184) and in Eq. (185), “mp” should be “1,mp”; see also [53]
[25]	In Eqs. (28), (29), (32) and (33), $nkT\lambda$ should be η_0 ; To the Fig. 6 caption, add “ $\lambda\dot{\gamma}^0 = \lambda\omega = 1$ ”; After Eq. (23), “which match those of Eq. (20), as they must.” should be “which, surprisingly, does not differ from the intercepts implied by the lower order result in Eq. (8).”; In Fig. 7, $\eta'(\lambda\omega, \lambda\dot{\gamma}^0)$ should be $\eta'(\lambda\omega, \lambda\dot{\gamma}^0) / nkT\lambda$; In Fig. 8, $\eta''(\lambda\omega, \lambda\dot{\gamma}^0) / \lambda\omega$ should be $\eta''(\lambda\omega, \lambda\dot{\gamma}^0) / nkT\lambda^2\omega$; Move errata following Ref. (80) to the ends of Refs. (27) and (76); In the last row of Table II, “storage” should be “loss and storage”.
[27]	In Eq. (91), η' should be η'' ; In caption to Fig. 3, “ $\Psi_1[P_2^2s_2]$ ” should be “cos3 ωt ” and “ $\Psi_2[P_2^0c_0, P_2^2c_2, \dots]$ ” should be “sin3 ωt ”; in Eq. (10), “38725” should be “38728”; On the left side of Eq. (31), “6” should be “–6” and “ $-\frac{1}{2}P_2^2s_2$ ” should be “ $\frac{1}{2}P_2^2s_2$ ”; In Eq. (32), “F” should be “–F” and “–f” should be “f”; in Eq. (33), “ $\frac{1}{12}$ ” should be “1”; In Eq. (37), G should be G(t); On the left side of Eq. (73), “ $\frac{\lambda}{2}$ ” should be “ $\frac{1}{4\pi} \frac{\lambda}{2}$ ”; In Eq. (60), “ $\int_{-\infty}^0$ ” should be “ $\int_{-\infty}^t$ ”; In the 4th line of Eq. (62), $\int_{-\infty}^0 \int_{-\infty}^s$ should be $\int_0^{\infty} \int_s^{\infty}$; In Eqs. (88) through (91), “+ $(\lambda\dot{\gamma}^0)^2$ ” should be “– $(\lambda\dot{\gamma}^0)^2$ ”; In Eqs. (86), (87), (90) and (91), “ $nkT\lambda$ ” should be “ η_0 ”; In captions to Figs. 7 and 8, “Coefficients” should be “Minus coefficients”.
[28]	Above Eqs. (14) and (25), “significant figures” should be “16 significant figures”
[29]	In Eq. (2.17), “ $NnkT$ ” should be “ $NnkT\lambda$ ”, the factor multiplying the term with cos3 ωt should be $2/7\lambda^2\omega^2$ and not $8/7\lambda^2\omega^2$, and $4(1-\frac{\epsilon}{5})$ in the final line should be $4(1+\frac{\epsilon}{5})$; in Eq. (2.23), the multiplicative factor should be $2/7\lambda^2\omega^2$ and not $8/7\lambda^2\omega^2$, and $s\lambda\omega$ should be $2\lambda\omega$; Eq. (2.29) is then reduced to 1/4 of the published result; in the second unnumbered equation following Eq. (A.3), $\sinh \sinh$ should be $\sinh$ and $\cosh$ should be $\cosh$.
[40]	In Eq. (12), both occurrences of “ τ ” should be “b”

[41]	In Eq. (5), the rightmost term should be multiplied by sin <i>not</i> , ordinate labels in Fig. 1 (a)–(d) should be Wi.
[60]	From 6th line of Introduction, delete “small-amplitude oscillatory shear”; in Eq. (6), “38725” should be “38728”; in Ref. [22], “[23]” should be “[21]”. In Eq. (27), “ $P_6^4c_4$ ” should be “ $\frac{7}{396}P_6^4c_4$ ”; In Eq. (32), “ $\frac{1}{63504}$ ” should be “ $\frac{-1}{63504}$ ”; In Eq. (33), “ $\frac{-1}{6\lambda} \frac{1}{432} \frac{3}{49} \frac{20}{7}$ ” should be “ $\frac{+1}{6\lambda} \frac{1}{432} \frac{3}{49} \frac{20}{7}$ ”; In Eq. (35), “ $\frac{-1}{74088}$ ” should be “ $\frac{+1}{74088}$ ”

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