QUEEN'S UNIVERSITY

POLYMER RESEARCH GROUP

19 Division Street, Kingston, ON, K7L 3N6 Canada

POLYMER ORIENTATION CONTRIBUTIONS IN LARGE-AMPLITUDE OSCILLATORY SHEAR FLOW

P.H. Gilbert¹ and A.J. Giacomin^{1,2,*}

¹Polymers Research Group
Chemical Engineering Department

²Mechanical and Materials Engineering Department
Queen's University
Kingston, ON K7L 3N6

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*Corresponding author (giacomin@chee.queensu.ca)

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ABSTRACT

Previously, we used a dilute suspension of rigid dumbbells as a model for the dynamics for polymeric liquids in large-amplitude oscillatory shear (LAOS) flow. We then use dumbbell orientation to explain fluid elasticity. We derived the expression for the polymer orientation distribution, and then we decomposed this function into its first five harmonics (the zeroth, first, second, third and fourth harmonics). We further separated the harmonics into their components, in-phase and out-of-phase with $\cos n\omega t$. In this work, we deepen our understanding of the relationship between the orientation distribution function and the shear stress and normal stress differences. We also investigate the components of orientation that make *no contribution at all* to measured rheological responses. Further, the larger the $\dot{\gamma}^0$ of the oscillatory shear flow, the greater the fraction of polymer that escapes rheological measurement. Our analysis focuses on the nonlinear viscoelastic regime, and specifically, where both $\lambda\omega$ and $\lambda\dot{\gamma}^0$ are unity. We learn that all orientation contributions to the normal stresses also contribute to the shear stress. The parts of the orientation distribution contributing to the shear stress take on the familiar peanut shapes of the total orientation distribution. The parts of the orientation distribution causing the normal stress differences are subsets of the part of the orientation distribution contributing to the shear stress.

Keywords: polymer dynamics, polymer orientation, large-amplitude oscillatory shear flow, Fourier-transform rheology, rigid-dumbbell suspension, structure-property relations

*Corresponding author (giacomin@queensu.ca).

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I. INTRODUCTION

The rigid dumbbell suspension is the simplest relevant model for polymer solutions and melts that captures their rheological behavior, at least qualitatively. By *relevant*, we at least mean that the model predicts higher harmonics of the shear stress in large-amplitude oscillatory shear flow (LAOS). A rigid dumbbell has fixed length and width, the two key geometric properties of a polymer molecule, and these confer orientation upon the polymer (see Figure 1). Polymer orientation contributes to the polymer rheology and is the source of rheological properties in rigid dumbbell suspensions. Oscillatory shear flow [1,2,3,4,5,6,7] is by far the most common experiment for probing polymer orientation [8], and when performed at large-amplitude ([9,10,11,12,13,14]; see also Section 2 of [15]), this test can be particularly informative. In this paper, we dissect molecular orientation to deepen our understanding of how this orientation causes the stress responses in LAOS. Table I classifies the literature on analytical solutions for orientation distributions in LAOS and compares the literature to this paper. By further contrast with this paper, see the detailed study of ψ in *small-amplitude* oscillatory shear flow of a rigid dumbbell suspension in *confined gaps* in Section 5. of [16] or Section 2.3.3 of [17].

Previously, we developed an expression for polymer orientation in LAOS from the diffusion equation, by expanding the orientation distribution in powers of shear rate amplitude [37,38,39,40,41,42]. We extended this work by decomposing the orientation distribution function into its first five (zeroth, first, second, third, fourth) harmonics [45,46]. By *decomposed*, we mean rewriting the orientation distribution as:

$$\psi(\theta, \phi, t) = \sum_{n=0}^{\infty} [\psi'_n \cos n\omega t + \psi''_n \sin n\omega t] \quad (1)$$

where ψ'_n and ψ''_n are the Fourier coefficients of the orientation distribution function respectively in-phase and out-of-phase with $\cos n\omega t$. These Fourier coefficients must not be confused with the orientation expansion coefficients, ψ_n , identified by Schmalzer and Giacomin [40,41,42], which are obtained from Legendre polynomials describing orientation. Subsequently, we found that the frequency content of each harmonic contributed orientation differently. Particularly, only the zeroth and second harmonics of ψ contribute orientation in the flow direction, whereas odd harmonics of ψ provide orientation away from both the flow direction and the y -axis (see coordinates defined in Figure 2). We also identified the second harmonic as the primary sculptor of the peanut shapes that arise in the overall orientation distribution, $\psi(\theta, \phi)$ [45].

This paper concerns itself with the orientation distribution function of a suspension of rigid dumbbells undergoing oscillatory shear flow [18,19], where $v_x(y, t)$, illustrated in Figure 2:

$$v_x = \dot{\gamma}^0 y \cos \omega t; v_y = v_z = 0 \quad (2)$$

All orientation distribution responses calculated herein refer to the cosinusoidal shear rate corresponding to Eq. (2) (see Section V of [20]). Our symbols, along

with their dimensions, are defined in Table II and Table III. Eq. (2) can be rewritten as:

$$\lambda\dot{\gamma}(t) = \lambda\dot{\gamma}^0 \cos \lambda\omega(t/\lambda) \quad (3)$$

where the dimensionless shear rate $\lambda\dot{\gamma}^0$ (the Weissenberg number) and the dimensionless frequency $\lambda\omega$ (the Deborah number) appear [18,19,22]. These dimensionless groups contain the characteristic time constant, λ , which, for the dilute suspension of rigid dumbbells in a Newtonian solvent of viscosity, η_s , is:

$$\lambda = \frac{\xi L^2}{12kT} = \frac{(3\pi\eta_s b)L^2}{12kT} \quad (4)$$

and for rigid multibead-rods:

$$\lambda_N = \frac{\xi L^2 N(N+1)}{72(N-1)kT} \quad (5)$$

where N is the number of equally-spaced identical beads on the rod of length L , T is absolute temperature, k is Boltzmann's constant, ξ is the bead friction factor given by Stokes' law, and b is the diameter of each of the beads of the dumbbell (see Figure 1). Eqs. (4) and (5) thus connect polymer molecular structure to the rheological properties; we call these *structure-property relations*.

We further restrict this paper to the periodic responses of the stresses and polymer orientation, reached several cycles after startup of oscillatory shear. We call this periodicity alternance (see Section 6. of [21]; [22]). This work is an extension of recent articles [37,38] wherein part of the orientation distribution is derived using the method of Bird and Armstrong [23] (see also Problem 11C.1 of [24]). Whereas, in previous work, we focused on the parts of orientation contributing to known rheological measurements (viscosity, shear stress and normal stress differences), this work deepens our understanding of which orientation components contribute to rheological responses, and which do not. We develop our understanding of the molecular motions by dissecting $\psi(\theta, \phi, t)$ into its contributing and noncontributing parts. We then illustrate these parts of $\psi(\theta, \phi, t)$ with colored orientation distribution diagrams and videography. For simplicity, we explore just one special case, $\lambda\dot{\gamma}^0 = \lambda\omega = 1$, where we know the rheological responses are far from both linear viscoelasticity and steady shear flow.

A. Orientation distribution function

For simple shear flow, the *diffusion equation* for the orientation distribution function $\psi(\theta, \phi, t)$ is given by (see Eq. 5.1 of [25], Eq. (14.4-1) of [26]):

$$6\lambda \frac{\partial \psi}{\partial t} = \left[\frac{1}{S} \frac{\partial}{\partial \theta} \left(S \frac{\partial \psi}{\partial \theta} \right) + \frac{1}{S^2} \frac{\partial^2 \psi}{\partial \phi^2} \right] - 6\lambda \dot{\gamma} \left[\frac{sc}{S} \frac{\partial}{\partial \theta} (S^2 C \psi) - \frac{\partial}{\partial \phi} (s^2 \psi) \right] \quad (6)$$

where $\lambda\dot{\gamma}$ is the dimensionless oscillatory shear rate [given by Eq. (3)]. In Eq. (6), we use the abbreviations $S \equiv \sin\theta$, $C \equiv \cos\theta$, $s \equiv \sin\phi$ and $c \equiv \cos\phi$. For ψ we adhere to the definition of Bird-Warner-Evans [25] which differs from that of

[24,26]. The relation between the distribution function used here, ψ , and the symbol used in the first [24] and second [26] editions of *Dynamics of Polymeric Liquids* is (see footnote 1 on p. 524 of [24]):

$$\psi_{\text{DPL,1e}} = \psi_{\text{DPL,2e}} = \psi \sin \theta \quad (7)$$

Also, whereas the distribution function is represented here as ψ , the second edition of *Dynamics of Polymeric Liquids* [26] uses f instead.

Dumbbell orientation is thus given in terms of the spherical coordinates θ and ϕ , defined in Figure 1. When the fluid is at rest, all orientations are equally likely, and ψ is equal to a normalization constant, $\frac{1}{4\pi}$, so that:

$$\int_0^{2\pi} \int_0^\pi \psi(\theta, \phi, t) \sin \theta d\theta d\phi = 1 \quad (8)$$

In other words, when the fluid is at rest, $\psi(\theta, \phi)$ is spherically symmetric.

Although the orientation distribution function for rigid dumbbell suspensions has not been measured directly, its implications for rheological behavior have been verified experimentally. For instance, reasonable qualitative behaviors for oscillatory shear flow are obtained for amplitudes both small [25] and large [37,38]. Although ψ has not been measured directly in LAOS, x-ray scattering has been used to shed light on ψ [27]. However, this x-ray scattering work is on platelet suspensions in LAOS [27], but there is none yet on systems relevant to rigid dumbbell suspensions in LAOS, such as polymeric liquids.

Schmalzer and Giacomini [40,41,42] recently solved Eq. (6) analytically for large-amplitude oscillatory shear flow, including all terms be they contributing to the rheological responses or not, using the method of Bird and Armstrong [23]. This solution has the form:

$$\psi(\theta, \phi, t) = \frac{1}{4\pi} \left[1 + (6\lambda\dot{\gamma}^0) \psi_1(\theta, \phi, t) + (6\lambda\dot{\gamma}^0)^2 \psi_2(\theta, \phi, t) + (6\lambda\dot{\gamma}^0)^3 \psi_3(\theta, \phi, t) + (6\lambda\dot{\gamma}^0)^4 \psi_4(\theta, \phi, t) + \dots \right] \quad (9)$$

where ψ_1 through ψ_4 are given by Eqs. (36), (44), (52) and (61) of [40]. Giacomini et al. [45] also undertook the Fourier decomposition of these expressions for ψ_1 through ψ_4 , putting the resulting Fourier coefficients under common denominators. We continue these analyses of the orientation distribution to find which orientation components contribute to the shear stress, which contribute to each normal stress difference, and which components do not contribute to these rheological responses at all. We construct plots for each orientation component outlined in Table IV and Table V to deepen our understanding of the relationship between orientation and the stress responses.

II. METHOD

To determine which elements of the orientation distribution function contribute to the shear stress response, we begin with Eq. (72) of [37]:

$$\frac{\tau_{yx} - \tau_{yx,s}}{nkT} = \frac{\lambda}{2} \left[\frac{d}{dt} \langle P_2^2 s_2 \rangle - \dot{\gamma}^0 \cos \omega t \langle 2(P_0^0 c_0 - P_2^0 c_0) - P_2^2 c_2 \rangle \right] \quad (10)$$

where:

$$P_0^0 = 1 \quad (11)$$

$$P_2^0 = \frac{1}{2}(3C^2 - 1) \quad (12)$$

$$P_2^2 = 3(1 - C^2) \quad (13)$$

Using Eq. (17) of [37] to replace the chevrons, $\langle \rangle$, in Eq. (10), we get Eq. (73) of [37]:

$$\frac{\tau_{yx} - \tau_{yx,s}}{nkT} = \frac{\lambda}{2} \frac{1}{4\pi} \left[\begin{aligned} & \frac{d}{dt} \int_0^{2\pi} \int_0^\pi P_2^2 s_2 \left[1 + (6\lambda\dot{\gamma}^0) \psi_1 + (6\lambda\dot{\gamma}^0)^2 \psi_2 + (6\lambda\dot{\gamma}^0)^3 \psi_3 + \dots \right] \sin\theta d\theta d\phi \\ & - 2\dot{\gamma}^0 \cos\omega t \int_0^{2\pi} \int_0^\pi (P_0^0 c_0 - P_2^0 c_0) \left[1 + (6\lambda\dot{\gamma}^0) \psi_1 + (6\lambda\dot{\gamma}^0)^2 \psi_2 + (6\lambda\dot{\gamma}^0)^3 \psi_3 + \dots \right] \sin\theta d\theta d\phi \\ & + \dot{\gamma}^0 \cos\omega t \int_0^{2\pi} \int_0^\pi P_2^2 c_2 \left[1 + (6\lambda\dot{\gamma}^0) \psi_1 + (6\lambda\dot{\gamma}^0)^2 \psi_2 + (6\lambda\dot{\gamma}^0)^3 \psi_3 + \dots \right] \sin\theta d\theta d\phi \end{aligned} \right] \quad (14)$$

where:

$$\psi_1(\theta, \phi, t) = \frac{1}{12} P_2^2 s_2(1; t) \quad (15)$$

$$\psi_2(\theta, \phi, t) = -\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0, 1; t) - \left(\frac{6}{7} P_4^0 c_0 - \frac{1}{28} P_4^4 c_4 \right) \left(\frac{7}{3}, 1; t \right) \right] \quad (16)$$

$$\begin{aligned} \psi_3(\theta, \phi, t) = & -\frac{1}{10584} \left[23(0, 0, 1; t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1; t \right) \right] P_2^2 s_2 \\ & - \frac{1}{432} \left[\frac{3}{49} \left(\frac{7}{3}, 0, 1; t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1; t \right) \right] P_4^2 s_2 \\ & + \frac{1}{432(14)} \left[\frac{1}{2} \left(\frac{7}{3}, 0, 1; t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1; t \right) \right] P_4^4 s_4 \\ & + \frac{1}{432(22)} \left[P_6^2 s_2 - \frac{1}{72} P_6^6 s_6 \right] \left(\frac{11}{3}, \frac{7}{3}, 1; t \right) \end{aligned} \quad (17)$$

$$\begin{aligned} \psi_4(\theta, \phi, t) = & -\frac{1}{74088} \left[\begin{aligned} & 23(0, 0, 0, 1; t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \\ & + 5 \left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t \right) + \frac{50}{11} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t \right) \end{aligned} \right] P_2^0 c_0 \\ & - \frac{1}{63504} \left[\begin{aligned} & 23(0, 0, 0, 1; t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \\ & + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t \right) \end{aligned} \right] P_2^2 c_2 \end{aligned} \quad (18)$$

and:

$$P_4^0 = \frac{1}{8}(35C^4 - 30C^2 + 3) \quad (19)$$

$$P_4^2 = \frac{15}{2}(1 - C^2)(7C^2 - 1) \quad (20)$$

$$P_4^4 = 105(1 - C^2)^2 \quad (21)$$

$$P_6^2 = \frac{105}{8}(1-C^2)(33C^4+18C^2+1) \quad (22)$$

$$P_6^6 = 10395(1-C^2)^3 \quad (23)$$

If we evaluate each Legendre polynomial, P_n^m , paired with a component of the orientation distribution function, ψ_p , we get:

$$\frac{1}{4\pi} \frac{d}{dt} \int_0^{2\pi} \int_0^\pi P_2^2 s_2 \left[\begin{aligned} &1 + (6\lambda\dot{\gamma}^0) \frac{1}{12} P_2^2 s_2(1;t) \\ &+ (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0,1;t) - \left(\frac{6}{7} P_4^0 c_0 - \frac{1}{28} P_4^4 c_4 \right) \left(\frac{7}{3}, 1;t \right) \right] \right) \\ &+ (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left[23(0,0,1;t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 s_2 \right. \\ &\quad \left. - \frac{1}{432} \left[\frac{3}{49} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_4^2 s_2 \right. \\ &\quad \left. + \frac{1}{432(14)} \left[\frac{1}{2} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_4^4 s_4 \right. \\ &\quad \left. + \frac{1}{432(22)} \left[P_6^2 s_2 - \frac{1}{72} P_6^6 s_6 \right] \left(\frac{11}{3}, \frac{7}{3}, 1;t \right) \right) \\ &+ (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{74088} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^0 c_0 \right. \\ &\quad \left. - \frac{1}{63504} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right. \\ &\quad \left. + 5 \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{50}{11} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right) P_2^0 c_0 \right. \\ &\quad \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right) P_2^2 c_2 \right) \end{aligned} \right] \sin \theta d\theta d\phi \quad (24)$$

$$\frac{1}{4\pi} \frac{d}{dt} \int_0^{2\pi} \int_0^\pi P_2^2 s_2 \left[\begin{aligned} &(6\lambda\dot{\gamma}^0) \frac{1}{12} P_2^2 s_2(1;t) \\ &+ (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left[23(0,0,1;t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 s_2 \right) \end{aligned} \right] \sin \theta d\theta d\phi \quad (25)$$

for the first integration. For the second integration, we get:

$$\begin{aligned}
& \left[1 + (6\lambda\gamma^0) \frac{1}{12} P_2^2 s_2(1;t) \right. \\
& + (6\lambda\gamma^0)^2 \left(-\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0,1;t) \right. \right. \\
& \left. \left. - \left(\frac{6}{7} P_4^0 c_0 - \frac{1}{28} P_4^4 c_4 \right) \left(\frac{7}{3}, 1;t \right) \right] \right) \\
& + (6\lambda\gamma^0)^3 \left(-\frac{1}{10584} \left[23(0,0,1;t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 s_2 \right. \\
& - \frac{1}{432} \left[\frac{3}{49} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_4^2 s_2 \\
& + \frac{1}{432(14)} \left[\frac{1}{2} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_4^4 s_4 \\
& \left. \left. + \frac{1}{432(22)} \left[P_6^2 s_2 - \frac{1}{72} P_6^6 s_6 \right] \left(\frac{11}{3}, \frac{7}{3}, 1;t \right) \right] \right. \\
& + (6\lambda\gamma^0)^4 \left(-\frac{1}{74088} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right. \right. \\
& \left. \left. + 5 \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{50}{11} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^0 c_0 \right. \\
& \left. - \frac{1}{63504} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right. \right. \\
& \left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right) \right] \sin \theta d\theta d\phi \quad (26)
\end{aligned}$$

$$\begin{aligned}
& \left[1 + (6\lambda\gamma^0)^2 \left(-\frac{1}{72} \left[\frac{6}{7} P_2^0 c_0 (0,1;t) \right] \right) \right. \\
& + (6\lambda\gamma^0)^4 \left(-\frac{1}{74088} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right. \right. \\
& \left. \left. + 5 \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{50}{11} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^0 c_0 \right) \right] \sin \theta d\theta d\phi \quad (27)
\end{aligned}$$

For the third integral, we find:

$$\begin{aligned}
& \frac{\dot{\gamma}^0 \cos \omega t}{4\pi} \int_0^{2\pi} \int_0^\pi P_2^2 c_2 \left[\begin{aligned} & 1 + (6\lambda\dot{\gamma}^0) \frac{1}{12} P_2^2 s_2(1;t) \\ & + (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0,1;t) \right. \right. \\ & \quad \left. \left. - \left(\frac{6}{7} P_4^0 c_0 - \frac{1}{28} P_4^4 c_4 \right) \left(\frac{7}{3}, 1;t \right) \right] \right) \\ & + (6\lambda\dot{\gamma}^0)^3 \left(\begin{aligned} & -\frac{1}{10584} \left[23(0,0,1;t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 s_2 \\ & -\frac{1}{432} \left[\frac{3}{49} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_4^2 s_2 \\ & +\frac{1}{432(14)} \left[\frac{1}{2} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_4^4 s_4 \\ & +\frac{1}{432(22)} \left[P_6^2 s_2 - \frac{1}{72} P_6^6 s_6 \right] \left(\frac{11}{3}, \frac{7}{3}, 1;t \right) \end{aligned} \right) \\ & + (6\lambda\dot{\gamma}^0)^4 \left(\begin{aligned} & -\frac{1}{74088} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right. \right. \\ & \quad \left. \left. + 5 \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{50}{11} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^0 c_0 \\ & -\frac{1}{63504} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right. \right. \\ & \quad \left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right) \end{aligned} \right] \sin \theta d\theta d\phi \quad (28) \\
& \frac{\dot{\gamma}^0 \cos \omega t}{4\pi} \int_0^{2\pi} \int_0^\pi P_2^2 c_2 \left[\begin{aligned} & (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} [(-P_2^2 c_2)(0,1;t)] \right) \\ & + (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{63504} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right. \right. \\ & \quad \left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right) \end{aligned} \right] \sin \theta d\theta d\phi \quad (29)
\end{aligned}$$

Substituting Eqs. (25), (27), and (29) into Eq. (14) yields:

$$\begin{aligned}
& \frac{\tau_{yx} - \tau_{yx,s}}{nkT} = \frac{\lambda}{2} \frac{1}{4\pi} \left[\begin{aligned} & \frac{d}{dt} \int_0^{2\pi} \int_0^\pi P_2^2 s_2 \left[\begin{aligned} & (6\lambda\dot{\gamma}^0) \frac{1}{12} P_2^2 s_2(1;t) \\ & + (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left[23(0,0,1;t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 s_2 \right) \end{aligned} \right] \sin \theta d\theta d\phi \\ & - 2\dot{\gamma}^0 \cos \omega t \int_0^{2\pi} \int_0^\pi (P_2^0 c_0 - P_2^2 c_2) \left[\begin{aligned} & 1 + (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[\frac{6}{7} P_2^0 c_0 (0,1;t) \right] \right) \\ & + (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{74088} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right. \right. \\ & \quad \left. \left. + 5 \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{50}{11} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^0 c_0 \right) \end{aligned} \right] \sin \theta d\theta d\phi \\ & + \dot{\gamma}^0 \cos \omega t \int_0^{2\pi} \int_0^\pi P_2^2 c_2 \left[\begin{aligned} & (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} [(-P_2^2 c_2)(0,1;t)] \right) \\ & + (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{63504} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right. \right. \\ & \quad \left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right) \end{aligned} \right] \sin \theta d\theta d\phi \end{aligned} \right] \quad (30)
\end{aligned}$$

the polymer contribution to the shear stress response, where each order of the orientation distribution expansion contributes. However, not all parts of each

orientation expansion term, Eqs. (15)-(18), appear in Eq. (30). We enumerate all orientation contributions to shear stress in cell (1,1) of Table IV.

We next turn our attention to the orientation contribution to the normal stress differences. The first normal stress difference from Eq. (12) of [23] is:

$$\frac{\tau_{xx} - \tau_{yy}}{nkT} = \frac{\lambda}{2} \frac{1}{4\pi} \left[\frac{d}{dt} \int_0^{2\pi} \int_0^\pi P_2^2 c_2 \left[1 + (6\lambda\dot{\gamma}^0) \psi_1 + (6\lambda\dot{\gamma}^0)^2 \psi_2 + (6\lambda\dot{\gamma}^0)^3 \psi_3 + \dots \right] \sin\theta d\theta d\phi \right. \\ \left. - \dot{\gamma}^0 \cos\omega t \int_0^{2\pi} \int_0^\pi P_2^2 s_2 \left[1 + (6\lambda\dot{\gamma}^0) \psi_1 + (6\lambda\dot{\gamma}^0)^2 \psi_2 + (6\lambda\dot{\gamma}^0)^3 \psi_3 + \dots \right] \sin\theta d\theta d\phi \right] \quad (31)$$

and replacing each term in the orientation distribution expansion, we find:

$$\frac{\lambda}{2} \frac{1}{4\pi} \frac{d}{dt} \int_0^{2\pi} \int_0^\pi P_2^2 c_2 \left[\begin{aligned} & 1 + (6\lambda\dot{\gamma}^0) \frac{1}{12} P_2^2 s_2(1;t) \\ & + (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0,1;t) \right. \right. \right. \\ & \quad \left. \left. \left. - \left(\frac{6}{7} P_4^0 c_0 - \frac{1}{28} P_4^4 c_4 \right) \left(\frac{7}{3}, 1;t \right) \right] \right) \right] \\ & + (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left[23(0,0,1;t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 s_2 \right. \\ & \quad - \frac{1}{432} \left[\frac{3}{49} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_4^2 s_2 \\ & \quad + \frac{1}{432(14)} \left[\frac{1}{2} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_4^4 s_4 \\ & \quad \left. + \frac{1}{432(22)} \left[P_6^2 s_2 - \frac{1}{72} P_6^6 s_6 \right] \left(\frac{11}{3}, \frac{7}{3}, 1;t \right) \right) \\ & + (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{74088} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right. \right. \\ & \quad \left. \left. + 5 \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{50}{11} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^0 c_0 \right. \right. \\ & \quad \left. \left. - \frac{1}{63504} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right. \right. \right. \\ & \quad \left. \left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right) \right] \right] \sin\theta d\theta d\phi \quad (32) \end{aligned}$$

$$\frac{\lambda}{2} \frac{1}{4\pi} \frac{d}{dt} \int_0^{2\pi} \int_0^\pi P_2^2 c_2 \left[\begin{aligned} & (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} [(-P_2^2 c_2)(0,1;t)] \right) \\ & + (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{63504} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right. \right. \right. \\ & \quad \left. \left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right) \right] \right] \sin\theta d\theta d\phi \quad (33) \end{aligned}$$

for the first integral and:

$$\begin{aligned}
& \left[\frac{\lambda}{2} \frac{1}{4\pi} -\dot{\gamma}^0 \cos \omega t \int_0^{2\pi} \int_0^\pi P_2^2 s_2 \right. \\
& \quad \left. + (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0,1;t) \right. \right. \right. \\
& \quad \left. \left. \left. - \left(\frac{6}{7} P_4^0 c_0 - \frac{1}{28} P_4^4 c_4 \right) \left(\frac{7}{3},1;t \right) \right] \right) \right. \\
& \quad \left. + (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left[23(0,0,1;t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 s_2 \right. \right. \\
& \quad \left. - \frac{1}{432} \left[\frac{3}{49} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_2^2 s_2 \right. \right. \\
& \quad \left. + \frac{1}{432(14)} \left[\frac{1}{2} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_4^4 s_4 \right. \right. \\
& \quad \left. + \frac{1}{432(22)} \left[P_6^2 s_2 - \frac{1}{72} P_6^6 s_6 \right] \left(\frac{11}{3}, \frac{7}{3}, 1;t \right) \right) \right. \\
& \quad \left. + (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{74088} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^0 c_0 \right. \right. \\
& \quad \left. + 5 \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{50}{11} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right) P_2^0 c_0 \right. \\
& \quad \left. - \frac{1}{63504} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right. \right. \\
& \quad \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right) P_2^2 c_2 \right) \right] \sin \theta d\theta d\phi \quad (34) \\
& \left. \frac{\lambda}{2} \frac{1}{4\pi} -\dot{\gamma}^0 \cos \omega t \int_0^{2\pi} \int_0^\pi P_2^2 s_2 \right. \\
& \quad \left. + (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left[23(0,0,1;t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 s_2 \right) \right] \sin \theta d\theta d\phi \quad (35)
\end{aligned}$$

for the second integral. Inserting Eqs. (33) and (35) into Eq. (31), we get the first normal stress difference:

$$\begin{aligned}
& \left[\frac{\tau_{xx} - \tau_{yy}}{nkT} = \frac{\lambda}{2} \frac{1}{4\pi} \right. \\
& \quad \left. \frac{d}{dt} \int_0^{2\pi} \int_0^\pi P_2^2 c_2 \right. \\
& \quad \left. + (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[(-P_2^2 c_2) (0,1;t) \right] \right) \right. \\
& \quad \left. + (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{63504} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right. \right. \right. \\
& \quad \left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right) \right] \sin \theta d\theta d\phi \quad (36) \\
& \quad \left. -\dot{\gamma}^0 \cos \omega t \int_0^{2\pi} \int_0^\pi P_2^2 s_2 \right. \\
& \quad \left. + (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left[23(0,0,1;t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 s_2 \right) \right] \sin \theta d\theta d\phi
\end{aligned}$$

All orders of the orientation distribution expansion, except the zeroth, contribute to the first normal stress difference, but not every part of each expansion term appears in the final expression. For instance, the first term in Eq. (18) is absent from the $(6\lambda\dot{\gamma}^0)^4$ coefficient of Eq. (36).

Repeating our analysis for the second normal stress difference, using Eq. (13) of [23], we get:

$$\frac{\tau_{yy} - \tau_{zz}}{nkT} = \lambda \frac{1}{4\pi} \left[\frac{d}{dt} \int_0^{2\pi} \int_0^\pi \left(-3P_2^0 - \frac{1}{2}P_2^2 c_2 \right) \left[1 + (6\lambda\dot{\gamma}^0)\psi_1 + (6\lambda\dot{\gamma}^0)^2\psi_2 + (6\lambda\dot{\gamma}^0)^3\psi_3 + \dots \right] \sin\theta d\theta d\phi \right] \quad (37)$$

which, after substitution of the orientation expansion terms, yields:

$$\frac{\tau_{yy} - \tau_{zz}}{nkT} = \lambda \frac{1}{4\pi} \left[\frac{d}{dt} \int_0^{2\pi} \int_0^\pi \left(-3P_2^0 - \frac{1}{2}P_2^2 c_2 \right) \left[\begin{aligned} & 1 + (6\lambda\dot{\gamma}^0) \frac{1}{12} P_2^2 s_2(1;t) \\ & + (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0,1;t) \right] \right. \right. \\ & \quad \left. \left. + (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left[23(0,0,1;t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 s_2 \right) \right. \right. \\ & \quad \left. \left. - \frac{1}{432} \left[\frac{3}{49} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_4^2 s_2 \right. \right. \\ & \quad \left. \left. + \frac{1}{432(14)} \left[\frac{1}{2} \left(\frac{7}{3}, 0, 1;t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1;t \right) \right] P_4^4 s_4 \right. \right. \\ & \quad \left. \left. + \frac{1}{432(22)} \left[P_6^2 s_2 - \frac{1}{72} P_6^6 s_6 \right] \left(\frac{11}{3}, \frac{7}{3}, 1;t \right) \right] \right. \right. \\ & \quad \left. \left. + (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{74088} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^0 c_0 \right. \right. \right. \right. \\ & \quad \left. \left. \left. - \frac{1}{63504} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right. \right. \right. \right. \\ & \quad \left. \left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] \right) \right] \sin\theta d\theta d\phi \right] \quad (38) \end{aligned}$$

and:

$$\frac{\tau_{yy} - \tau_{zz}}{nkT} = \lambda \frac{1}{4\pi} \left[\frac{d}{dt} \int_0^{2\pi} \int_0^\pi \left(-3P_2^0 - \frac{1}{2}P_2^2 c_2 \right) \left[\begin{aligned} & (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0,1;t) \right] \right) \right. \\ & \quad \left. + (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{74088} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^0 c_0 \right. \right. \right. \\ & \quad \left. \left. \left. - \frac{1}{63504} \left[23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \right] P_2^2 c_2 \right. \right. \right. \\ & \quad \left. \left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \right] \right) \right] \sin\theta d\theta d\phi \right] \quad (39) \end{aligned}$$

and thus, only the second and fourth orders of the orientation distribution expansion contribute to the second normal stress difference.

Though each orientation expansion term in Eqs. (15)-(18) contributes to at least one of the stress responses, portions of the second and third orientation expansion coefficients in Eqs. (16)-(17) do not contribute to any of the stresses. Specifically, there are no contributions to stress from the terms including: P_4^0 , P_4^2 , P_4^4 , P_6^2 or P_6^6 . There are therefore parts of the orientation distribution that escape rheological measurement. Table IV and Table V summarize the orientation contributions to the shear stress and normal stress differences.

To further understand how orientation contributes to the stresses, we develop plots of the orientation distribution function where only the parts appearing in each cell of Table IV and Table V are included. We thus construct the orientation distribution that contributes to the shear stress (see cell (1,1) of Table IV, cell (1,4)

of Table V, and Figure 3) by inserting the spherical harmonics remaining in Eq. (30) into Eq. (9) to get:

$$\psi_{ss}(\theta, \phi, t) = \frac{1}{4\pi} \left[\begin{aligned} &1 + (6\lambda\dot{\gamma}^0) \frac{1}{12} P_2^2 s_2(1; t) \\ &+ (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0, 1; t) \right] \right) \\ &+ (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left[23(0, 0, 1; t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1; t \right) \right] P_2^2 s_2 \right) \\ &+ (6\lambda\dot{\gamma}^0)^4 \left(\begin{aligned} &\left(-\frac{1}{74088} \left[23(0, 0, 0, 1; t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \right] P_2^0 c_0 \right. \\ &\left. + \frac{1}{63504} \left[23(0, 0, 0, 1; t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \right. \right. \\ &\left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t \right) \right] P_2^2 c_2 \right) \end{aligned} \right) \end{aligned} \right] \quad (40)$$

We next substitute the expressions for the paren functions (APPENDIX A of [42]):

$$(1; t) = \frac{\cos \omega t + \lambda \omega \sin \omega t}{(1 + W)} \quad (41)$$

$$(1; t) \cos \omega t = \frac{\cos 2\omega t + \lambda \omega \sin 2\omega t}{2(1 + W)} + \frac{1}{2(1 + W)} \quad (42)$$

$$(0, 1; t) = \frac{[1 - 2W] \cos 2\omega t + 3\lambda \omega \sin 2\omega t}{2(1 + 4W)(1 + W)} + \frac{1}{2(1 + W)} \quad (43)$$

$$(0, 1; t) \cos \omega t = \frac{[1 - 2W](\cos \omega t + \cos 3\omega t) + 3\lambda \omega (\sin \omega t + \sin 3\omega t)}{4(1 + 4W)(1 + W)} + \frac{\cos \omega t}{2(1 + W)} \quad (44)$$

$$\left(\frac{2}{3}, 1; t\right) = \frac{3(10 - 6W) \cos 2\omega t + 48\lambda \omega \sin 2\omega t}{16(25 + 9W)(1 + W)} + \frac{3}{20(1 + W)} \quad (45)$$

$$(0, 0, 1; t) = \frac{(1 - 11W) \cos 3\omega t + 6(1 - W) \lambda \omega \sin 3\omega t}{4(1 + 9W)(1 + 4W)(1 + W)} \quad (46)$$

$$\begin{aligned} &+ \frac{3 \cos \omega t + 6 \lambda \omega \sin \omega t}{4(1 + 4W)(1 + W)} \\ \left(-\frac{2}{3}, \frac{2}{3}, 1; t\right) &= \frac{3(5 - 27W) \cos 3\omega t + 3(23 - 9W) \lambda \omega \sin 3\omega t}{8(1 + 9W)(25 + 9W)(1 + W)} \\ &+ \frac{3(75 - 37W) \cos \omega t + 3(3W + 115) \lambda \omega \sin \omega t}{40(25 + 9W)(1 + W)^2} \end{aligned} \quad (47)$$

$$\begin{aligned} \left(\frac{2}{3}, 0, 1; t\right) &= \frac{3(10 - 47W) \cos 3\omega t + 9(13 - 6W) \lambda \omega \sin 3\omega t}{4(1 + 4W)(100 + 81W)(1 + W)} \\ &+ \frac{9(10 + 17W) \cos \omega t + 9(13 + 6W) \lambda \omega \sin \omega t}{4(1 + 4W)(100 + 9W)(1 + W)} \end{aligned} \quad (48)$$

$$\begin{aligned} \left(0, \frac{2}{3}, 1; t\right) &= \frac{18(25 - 51W) \cos 3\omega t + 9(125 - 27W) \lambda \omega \sin 3\omega t}{8(25 + 9W)(100 + 81W)(1 + W)} \\ &+ \frac{270(25 - 3W) \cos \omega t + 9(625 + 9W) \lambda \omega \sin \omega t}{40(1 + 4W)(100 + 9W)(1 + W)} \end{aligned} \quad (49)$$

$$\left(\frac{11}{3}, \frac{7}{3}, 1; t\right) = \frac{15(7-9W)\cos 3\omega t + 3(71-9W)\lambda\omega \sin 3\omega t}{8(25+9W)(49+9W)(1+W)} \quad (50)$$

$$+ \frac{3(525-19W)\cos \omega t + 3(355+3W)\lambda\omega \sin \omega t}{40(25+9W)(49+W)(1+W)}$$

$$(0, 0, 0, 1; t) = \frac{(1-35W+24W^2)\cos 4\omega t + (10-50W)\lambda\omega \sin 4\omega t}{8(1+16W)(1+9W)(1+4W)(1+W)} \quad (51)$$

$$+ \frac{(4-13W-32W^2-159W^3)\cos 2\omega t + 2(10+26W+61W^2-27W^3)\lambda\omega \sin 2\omega t}{8(1+4W)(1+9W)(1+W)^3}$$

$$+ \frac{3}{8(1+4W)(1+W)}$$

$$(0, -\frac{7}{3}, \frac{7}{3}, 1; t) = 3 \frac{(5-119W+36W^2)\cos 4\omega t + (43-117W)\lambda\omega \sin 4\omega t}{4(1+16W)(1+9W)(100+36W)(1+W)} \quad (52)$$

$$+ 3 \frac{\left[\begin{aligned} &(100+243W-2289W^2-5915W^3-1179W^4)\cos 2\omega t \\ &+ 2(215+1447W+1934W^2-693W^3-243W^4)\lambda\omega \sin 2\omega t \end{aligned} \right]}{20(1+4W)(1+9W)(100+36W)(1+W)^3}$$

$$+ \frac{3(75-37W)}{20(1+W)^2(100+36W)}$$

$$(-\frac{7}{3}, \frac{7}{3}, 0, 1; t) = 3 \frac{(10-203W+72W^2)\cos 4\omega t + (79-206W)\lambda\omega \sin 4\omega t}{8(1+16W)(100+81W)(1+4W)(1+W)} \quad (53)$$

$$+ 3 \frac{\left[\begin{aligned} &(4000-5680W-10262W^2-38031W^3-30780W^4)\cos 2\omega t \\ &+ (15800+35050W+63881W^2+32130W^3-5832W^4)\lambda\omega \sin 2\omega t \end{aligned} \right]}{8(1+16W)(100+81W)(1+4W)(1+W)^2(100+9W)}$$

$$+ \frac{9(10+17W)}{8(100+9W)(1+4W)(1+W)}$$

$$(-\frac{7}{3}, 0, \frac{7}{3}, 1; t) = 9 \frac{2(25-301W+54W^2)\cos 4\omega t + 5(65-87W)\lambda\omega \sin 4\omega t}{4(1+16W)(100+81W)(100+36W)(1+W)} \quad (54)$$

$$+ 9 \frac{\left[\begin{aligned} &(100000-72000W-475930W^2-330813W^3-62451W^4)\cos 2\omega t \\ &+ (325000+622150W+269979W^2-4725W^3-13122W^4)\lambda\omega \sin 2\omega t \end{aligned} \right]}{20(1+4W)(100+81W)(100+36W)(1+W)^2(100+9W)}$$

$$+ \frac{27(25-3W)}{2(100+9W)(100+36W)(1+W)}$$

and insert the definitions of each Legendre polynomial [Eqs. (11)-(13)] into Eq. (40) to get:

$$\psi_{ss}(\theta, \phi, t) = \frac{1}{4\pi} \left[\begin{aligned} &1 + (6\lambda\dot{\gamma}^0) \frac{1}{12} P_2^2 s_2(1; t) \\ &+ (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0, 1; t) \right] \right) \\ &+ (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left[23(0, 0, 1; t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1; t \right) \right] P_2^2 s_2 \right) \\ &+ (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{74088} \left[23(0, 0, 0, 1; t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \right. \right. \\ &\quad \left. \left. + 5 \left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t \right) + \frac{50}{11} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t \right) \right] P_2^0 c_0 \right. \\ &\quad \left. -\frac{1}{63504} \left[23(0, 0, 0, 1; t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \right. \right. \\ &\quad \left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t \right) \right] P_2^2 c_2 \right] \end{aligned} \right) \quad (55)$$

$$\psi_{ss}(\theta, \phi, t) = \frac{1}{4\pi} \left[\begin{aligned} &1 + (6\lambda\dot{\gamma}^0) \frac{1}{12} 3(1 - C^2) s_2 \left(\frac{\cos \omega t + \lambda \omega \sin \omega t}{(1 + W)} \right) \\ &+ (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[\left(\frac{6}{7} \frac{1}{2} (3C^2 - 1) - 3(1 - C^2) c_2 \right) (0, 1; t) \right] \right) \\ &+ (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left(\begin{aligned} &23 \left[\frac{(1 - 11W) \cos 3\omega t + 6(1 - W) \lambda \omega \sin 3\omega t}{4(1 + 9W)(1 + 4W)(1 + W)} \right. \right. \\ &\quad \left. \left. + \frac{3 \cos \omega t + 6 \lambda \omega \sin \omega t}{4(1 + 4W)(1 + W)} \right] \right. \right. \\ &\quad \left. \left. + 12 \left[\frac{3(5 - 27W) \cos 3\omega t + 3(23 - 9W) \lambda \omega \sin 3\omega t}{8(1 + 9W)(25 + 9W)(1 + W)} \right. \right. \right. \\ &\quad \left. \left. \left. + \frac{3(75 - 37W) \cos \omega t + 3(3W + 115) \lambda \omega \sin \omega t}{40(25 + 9W)(1 + W)^2} \right] \right) 3(1 - C^2) s_2 \right) \end{aligned} \right) \quad (56) \\ &+ (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{74088} \left[23(0, 0, 0, 1; t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \right. \right. \\ &\quad \left. \left. + 5 \left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t \right) + \frac{50}{11} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t \right) \right] \frac{1}{2} (3C^2 - 1) \right. \\ &\quad \left. -\frac{1}{63504} \left[23(0, 0, 0, 1; t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \right. \right. \\ &\quad \left. \left. + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t \right) \right] 3(1 - C^2) c_2 \right) \end{aligned} \right)$$

$$\begin{aligned}
\psi_{ss}(\theta, \phi, t) = \frac{1}{4\pi} & \left[\begin{aligned} & 1 + (6\lambda\dot{\gamma}^0) \frac{1}{4}(1-C^2)s_2 \left(\frac{\cos\omega t + \lambda\omega \sin\omega t}{(1+W)} \right) \\ & + (6\lambda\dot{\gamma}^0)^2 \left(-\frac{1}{72} \left[\left(\frac{6}{7} \frac{1}{2} (3C^2-1) - 3(1-C^2)c_2 \right) (0,1;t) \right] \right) \\ & + (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left(\begin{aligned} & \left[\frac{23(1-11W)\cos 3\omega t + 138(1-W)\lambda\omega \sin 3\omega t}{4(1+9W)(1+4W)(1+W)} \right. \right. \\ & \left. \left. + \frac{36(5-27W)\cos 3\omega t + 36(23-9W)\lambda\omega \sin 3\omega t}{8(1+9W)(25+9W)(1+W)} \right] \right. \right. \\ & \left. \left. + \left[\frac{36(75-37W)(1+4W)\cos\omega t + 36(1+4W)(3W+115)\lambda\omega \sin\omega t}{40(1+4W)(25+9W)(1+W)^2} \right. \right. \right. \\ & \left. \left. \left. + \frac{690(1+W)(25+9W)\cos\omega t + 1380(1+W)(25+9W)\lambda\omega \sin\omega t}{40(1+4W)(25+9W)(1+W)^2} \right] \right) \right) 3(1-C^2)s_2 \\ & + (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{74088} \left[\begin{aligned} & 23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \end{aligned} \right] \frac{1}{2}(3C^2-1) \\ & -\frac{1}{63504} \left[\begin{aligned} & 23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \\ & + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \end{aligned} \right] 3(1-C^2)c_2 \end{aligned} \right) \right] \quad (57)
\end{aligned}$$

$$\begin{aligned}
\psi_{ss}(\theta, \phi, t) = \frac{1}{4\pi} & \left[\begin{aligned} & 1 + (6\lambda\dot{\gamma}^0) \left[\frac{1}{4}(1-C^2)s_2 \left(\frac{\cos\omega t + \lambda\omega \sin\omega t}{(1+W)} \right) \right] \\ & - \frac{3}{144} (6\lambda\dot{\gamma}^0)^2 \left(\frac{1}{7}(3C^2-1) - (1-C^2)c_2 \right) \left(\frac{(1-2W)\cos 2\omega t + 3\lambda\omega \sin 2\omega t}{(1+4W)(1+W)} + \frac{1}{(1+W)} \right) \\ & + (6\lambda\dot{\gamma}^0)^3 \left(-\frac{1}{10584} \left(\begin{aligned} & \left[\frac{23(1-11W)\cos 3\omega t + 138(1-W)\lambda\omega \sin 3\omega t}{4(1+9W)(1+4W)(1+W)} \right. \right. \\ & \left. \left. + \frac{36(5-27W)\cos 3\omega t + 36(23-9W)\lambda\omega \sin 3\omega t}{8(1+9W)(25+9W)(1+W)} \right] \right. \right. \\ & \left. \left. + \left[\frac{19950 + 59646W + 882W^2}{40(1+4W)(25+9W)(1+W)^2} \cos\omega t \right. \right. \right. \\ & \left. \left. \left. + \frac{36(1+4W)(3W+115) + 1380(1+W)(25+9W)}{40(1+4W)(25+9W)(1+W)^2} \lambda\omega \sin\omega t \right] \right) \right) 3(1-C^2)s_2 \\ & + (6\lambda\dot{\gamma}^0)^4 \left(-\frac{1}{74088} \left[\begin{aligned} & 23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \end{aligned} \right] \frac{1}{2}(3C^2-1) \\ & -\frac{1}{63504} \left[\begin{aligned} & 23(0,0,0,1;t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1;t \right) \\ & + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1;t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1;t \right) \end{aligned} \right] 3(1-C^2)c_2 \end{aligned} \right) \right] \quad (58)
\end{aligned}$$

$$\begin{aligned}
\psi_{ss}(\theta, \phi, t) = \frac{1}{4\pi} & \left[1 + (6\lambda\dot{\gamma}^0) \left\{ \frac{1}{4}(1-C^2)s_2 \left(\frac{\cos \omega t + \lambda\omega \sin \omega t}{1+W} \right) \right. \right. \\
& - \frac{1}{72}(6\lambda\dot{\gamma}^0)^2 \left\{ \left[\frac{\frac{24}{7}(3C^2-1)(1-2W)(25+9W)}{-24(1-C^2)(1-2W)(25+9W)c_2} \right] \cos 2\omega t \right. \\
& + \left[\frac{\frac{9}{7}(3C^2-1)(25+9W)}{-9(1-C^2)(25+9W)c_2} \right] \lambda\omega \sin 2\omega t \\
& + \left. \left. \left[\frac{\frac{30}{7}(3C^2-1)-30(1-C^2)c_2}{20(1+W)} \right] \right\} \right. \\
& + (6\lambda\dot{\gamma}^0)^3 \left\{ \left[\frac{(603W^2+892W-95)s_2(1-C^2)}{2016(W+1)(4W+1)(9W+1)(9W+25)} \right] \cos 3\omega t \right. \\
& + \left[\frac{(45W^2+17W-92)s_2(1-C^2)}{336(W+1)(4W+1)(9W+1)(9W+25)} \right] \lambda\omega \sin 3\omega t \\
& + \left[\frac{-(21W^2+784W+475)s_2(1-C^2)}{3360(W+1)^2(1+4W)(9W+25)} \right] \cos \omega t \\
& + \left. \left. \left[\frac{-(153W^2+757W+460)s_2(1-C^2)}{1680(W+1)^2(4W+1)(9W+25)} \right] \lambda\omega \sin \omega t \right\} \right. \\
& + (6\lambda\dot{\gamma}^0)^4 \left\{ \left[\frac{5(216W^3-201W^2-497W+16)(3C^2-1)}{133056(W+1)(4W+1)(9W+1)(9W+25)(16W+1)} \right] \cos 4\omega t \right. \\
& + \left[\frac{5(29160W^4-11556W^3-107889W^2-63124W+2225)(1-C^2)c_2}{8064(W+1)(4W+1)(9W+1)(9W+25)(16W+1)(81W+100)} \right] \lambda\omega \sin 4\omega t \\
& + \left[\frac{(18549162W^3+21819195W^2+7990963W-3933950)(3C^2-1)}{6519744(W+1)(4W+1)(9W+1)(9W+25)(16W+1)(81W+100)} \right] \cos 2\omega t \\
& + \left[\frac{836244W^3+1458315W^2+392806W-211025}{16128(1+W)(1+4W)(1+9W)(1+16W)(25+9W)(100+81W)} \right] \lambda\omega \sin 2\omega t \\
& + \left[\frac{(27917501148W^7+395970769863W^6+1359686945763W^5}{+1401100327677W^4+389057165809W^3+45722628300W^2} \right] (3C^2-1) \\
& + \left[\frac{-22856340000W-1568000000}{6519744(W+1)^3(4W+1)(9W+1)(9W+25)(9W+100)(16W+1)(81W+100)} \right] \cos 2\omega t \\
& + \left[\frac{(506797884W^7+6027824709W^6+1947775889W^5}{+19601882811W^4+5251665787W^3+424722600W^2} \right] (1-C^2)c_2 \\
& + \left[\frac{-340140000W-23000000}{40320(W+1)^3(4W+1)(9W+1)(9W+25)(16W+1)(81W+100)} \right] \cos 2\omega t \\
& + \left[\frac{(4554095076W^7+54922330017W^6+62656044453W^5}{-366082940052W^4-649050340979W^3-299118407355W^2} \right] (3C^2-1) \\
& + \left[\frac{-75936742500W-3787700000}{325987320(W+1)^3(4W+1)(9W+1)(9W+25)(9W+100)(16W+1)(81W+100)} \right] \lambda\omega \sin 2\omega t \\
& + \left[\frac{(73929348W^7+725310531W^6+487357884W^5}{-5792477481W^4-9601088132W^3-4427460420W^2} \right] (1-C^2)c_2 \\
& + \left[\frac{-1076466750W-53975000}{20160(W+1)^3(4W+1)(9W+1)(9W+25)(9W+100)(16W+1)(81W+100)} \right] \lambda\omega \sin 2\omega t \\
& - \left[\frac{(1161W^3+14769W^2+74200W+40000)(3C^2-1)}{221760(W+1)^2(4W+1)(9W+25)(9W+100)} \right] \\
& + \left. \left. \left[\frac{(1107W^3+19303W^2+60050W+28750)(1-C^2)c_2}{6720(W+1)^2(4W+1)(9W+25)(9W+100)} \right] \right\} \right]
\end{aligned} \tag{59}$$

which is the portion of the orientation distribution function that is contributing to the shear stress. We call ψ_{ss} the *shear stress orientation distribution*. We repeat this process in Section III to explore each part of orientation and how each of these parts contributes to each rheological response or not.

III. ORIENTATION RESULTS

Whereas the orientation distribution function provides all of the information necessary to determine the polymer stress responses, not all of the information contained in the polymer orientation is captured by either the shear stress or the normal stress differences. We deepen our understanding of orientation contribution and non-contribution to polymer rheology by exploring each row of Table IV and Table V, which detail the contributing and noncontributing parts of orientation. To better visualize these orientation components responsible for each stress response, we complete our analysis by developing three-dimensional orientation distributions for each cell in Table IV and Table V (Figure 3 - Figure 11). In each figure, high probability orientations are represented in red, and low, in blue. Our analysis focuses on orientation in the nonlinear viscoelastic regime, where both $\lambda\omega$ and $\lambda\dot{\gamma}^0$ are high, and specifically, where $\lambda\omega = \lambda\dot{\gamma}^0 = 1$.

A. Contribution to shear stress

Eq. (59) gives the orientation contribution to the shear stress for a rigid dumbbell suspension in large-amplitude oscillatory shear flow. Whereas the shear stress response is odd, the orientation is neither even nor odd. Furthermore, whereas the shear stress response only includes odd harmonics (see Eq. (20) of [47]), both odd and even harmonics of the orientation distribution function contribute to this shear stress. For instance, the third harmonic of the shear stress is caused by more than just the third harmonic of the orientation. Specifically, whereas the Fourier expansion of the shear stress contains only the *zeroth, second and fourth* powers of $\lambda\dot{\gamma}^0$ (see Eq. (20) of [47]), its cause depends on the *all* orders of the orientation distribution expansion.

We create Figure 3 from Eq. (59) to show the orientations causing the shear stress (see cell (1,1) of Table IV, cell (1,4) of Table V). The shear stress orientation distribution adopts either an egg or a peanut shape during an alternant cycle. These distribution shapes are reminiscent of those found in Figure 28 of [40], which is the total orientation distribution including all known orientation expansion terms $(1, \psi_1, \psi_2, \psi_3, \psi_4)$. The peanuts in Figure 3 are more bulbous than those in Figure 28 of [40] suggesting that the orientations contributing to the shear stress are more varied than those contributing to the total orientation distribution. Even for the egg shapes in Figure 3, corresponding to flow reversals ($\omega t = \frac{3\pi n}{4}$), a broader range of orientations contribute to the shear stress than to the overall orientation distribution (Figure 28 of [40]). Our video included with Figure 3 shows the transition from peanut to egg and back as the flow reverses.

If we isolate the orientation contributions to the shear stress that also contribute to the first normal stress difference (see Figure 4, cells (1,2) and (2,1-2) of Table IV and cell (2,4) of Table V), we obtain butterfly shapes where each wing tip is of equal magnitude. Comparing Figure 4 with Figure 3, we see that the butterfly wings occur both along and perpendicular to the peanuts and eggs of Figure 3. Since the perpendicular wings of Figure 4 appear to be absent from Figure 3, the contribution of orientation to both the shear stress and the first normal stress difference is much smaller than the full contribution to the shear

stress. Our video included with Figure 4 shows that the butterflies do not deform but just rock back and forth as the flow direction alternates.

Repeating this analysis for orientations contributing to both the shear stress and the second normal stress difference (Figure 5, see cells (1,3), (3,1) and (3,3) of Table IV and cell (3,4) of Table V), we discover lobes of alternating length. The *primary* orientations are along the flow direction (x – axis) and the *secondary* orientations occur along the y – axis. During each cycle, the secondary lobes shorten and lengthen while the primary lobes neither move nor deform (see our video included with Figure 5). The orientations in Figure 5 are not seen in the full shear stress orientation distribution of Figure 3. However, the secondary lobe variations of Figure 5 seem to explain the variation in peanut and egg girth along the y – axis in Figure 3, with the longer lobes explaining the egg shapes.

Inverting the discussion in the previous two paragraphs, we next isolate the shear stress orientation components that make no contribution to the first normal stress difference (see cell (1,2) of Table V). Figure 8 illustrates a unique orientation distribution that looks much like a car wheel, where the orientation is primarily in the xy – plane. Less important orientation contributions exist along the blue wheel rims. This suggests that the peanut shapes of Figure 3 are mostly caused by the parts of the shear stress orientation distribution that also contribute to the first normal stress difference. This finding is further supported by the peanut-shaped distributions of Figure 9, where only shear stress orientations making no contribution to the second normal stress difference are included (see cell (1,3) of Table V). When the contributions to the first normal stress difference (Figure 4) are reincorporated into the shear stress orientation distribution of Figure 8, we recover the peanut and egg shapes of Figure 3.

The shear stress orientation distribution (Figure 3) gives orientations oscillating in the directions that Figure 28 of [40] would lead us to expect. However, orientations associated with the first normal stress difference are necessary for creating the familiar orientation distribution shapes that are present in the total orientation distribution (Figure 28 of [40]). For this, we next deepen our understanding of the first normal stress difference orientation distribution.

B. Contribution to first normal stress difference

Inserting the spherical harmonics remaining in Eq. (36) into Eq. (9), we get the part of the orientation distribution function causing the first normal stress difference:

$$\psi_{N1}(\theta, \phi, t) = \frac{1}{4\pi} \left[\begin{aligned} & (6\lambda\dot{\gamma}^0) \frac{1}{12} P_2^2 s_2(1; t) \\ & + \frac{1}{72} (6\lambda\dot{\gamma}^0)^2 P_2^2 c_2(0, 1; t) \\ & - \frac{1}{10584} (6\lambda\dot{\gamma}^0)^3 \left[23(0, 0, 1; t) + 12 \left(-\frac{7}{3}, \frac{7}{3}, 1; t \right) \right] P_2^2 s_2 \\ & - \frac{1}{63504} (6\lambda\dot{\gamma}^0)^4 \left[\begin{aligned} & 23(0, 0, 0, 1; t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \\ & + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t \right) \end{aligned} \right] P_2^2 c_2 \end{aligned} \right] \quad (60)$$

which depends on the first through fourth orders of the orientation distribution expansion. We call ψ_{N1} the *first normal stress difference orientation distribution*.

From Eq. (60) we learn that, whereas the Fourier expansion of the first normal stress difference contains only the *second and fourth* powers of $\lambda\dot{\gamma}^0$ (see Eq. (13) of [38]), its cause depends on the *first through fourth* orders of the orientation distribution expansion.

Inserting the paren functions from Eqs. (41)-(54) and the Legendre polynomials from Eqs. (11)-(13) into Eq. (60), we get:

$$\psi_{N1}(\theta, \phi, t) = \frac{1}{4\pi} \left[\begin{aligned} & \frac{1}{4} (6\lambda\dot{\gamma}^0) (1-C^2) s_2 \frac{\cos \omega t + \lambda \omega \sin \omega t}{(1+W)} \\ & + \frac{1}{24} (6\lambda\dot{\gamma}^0)^2 (1-C^2) c_2 \left(\frac{[1-2W] \cos 2\omega t + 3\lambda \omega \sin 2\omega t}{2(1+4W)(1+W)} + \frac{1}{2(1+W)} \right) \\ & + \frac{1}{10080} (6\lambda\dot{\gamma}^0)^3 \left[\begin{aligned} & \frac{5}{(1+9W)(1+4W)(1+W)(25+9W)} \cos 3\omega t \\ & + 30 \frac{45W^2 + 17W - 92}{(1+9W)(1+4W)(1+W)(25+9W)} \lambda \omega \sin 3\omega t \\ & - 3 \frac{21W^2 + 784W + 475}{(1+4W)(1+W)^2(25+9W)} \cos \omega t \\ & - 6 \frac{153W^2 + 757W + 460}{(1+4W)(1+W)^2(25+9W)} \lambda \omega \sin \omega t \end{aligned} \right] (1-C^2) s_2 \\ & + \frac{1}{80640} (6\lambda\dot{\gamma}^0)^4 \left[\begin{aligned} & -50 \frac{29160W^4 - 11556W^3 - 107889W^2 - 63124W + 2225}{(1+W)(1+16W)(1+9W)(1+4W)(25+9W)(100+81W)} \cos 4\omega t \\ & + 5 \frac{836244W^3 + 1458315W^2 + 392806W - 211025}{(1+W)(1+4W)(1+9W)(1+16W)(25+9W)(100+81W)} \lambda \omega \sin 4\omega t \\ & + 2 \frac{(506797884W^7 + 6027824709W^6 + 19477775889W^5 + 19601882811W^4 + 5251665787W^3 + 424722600W^2 - 340140000W - 23000000)}{(1+16W)(1+9W)(1+4W)(1+W)^3(25+9W)(100+81W)(100+9W)} \cos 2\omega t \\ & + 4 \frac{(-9601088132W^3 - 4427460420W^2 - 1076466750W - 53975000)}{(1+16W)(1+9W)(1+4W)(1+W)^3(25+9W)(100+81W)(100+9W)} \lambda \omega \sin 2\omega t \\ & - 12 \frac{1107W^3 + 19303W^2 + 60050W + 28750}{(100+9W)(25+9W)(1+W)^2(1+4W)} \end{aligned} \right] (1-C^2) c_2 \end{aligned} \right] \quad (61)$$

which includes *all orientation harmonics* (zeroth through fourth). This differs from the Fourier analysis of the first normal stress difference, which includes only *even*

harmonics (zeroth, second, and fourth) (see Eq. (13) of [39]). In other words, the parities of ψ_{N_1} and N_1 differ. Whereas N_1 is even, ψ_{N_1} is neither even nor odd.

From Eq. (61), we generate Figure 4, the 3D orientation distribution contributing to the first normal stress difference (see cells (1,2) and (2,1-2) of Table IV and cell (2,4) of Table V). The orientation distributions take the form of butterflies with orientations both along and perpendicular to the peanuts of Figure 3. All of the orientations that contribute to the first normal stress difference also contribute to the shear stress. Thus, Figure 4 at once depicts the orientation distribution for the first normal stress difference, and also depicts the orientations contributing to both the first normal stress difference and the shear stress.

If we now focus on the parts of orientation contributing to both of the normal stress differences (see cells (2,3) and (3,2) of Table IV), we see that the butterflies orient themselves along the x – and y – axes and remain visibly stagnant throughout an alternant cycle (see Figure 6 and corresponding video). These are molecular motions participating in both N_1 and N_2 . Thus, the time constant parts of the $(\lambda\dot{\gamma}^0)^4$ terms in Eqs. (61) and (63) dominate the orientation.

We next obtain Figure 10 by examining the inverse condition to the previous paragraph, where we now exclude orientations shared by the normal stresses but include all other contributions to the first normal stress orientation (see cell (2,3) of Table IV). The butterflies now tilt away from the axes with wing tips at $\frac{\pi}{4}$. As we observed in Figure 6, the butterflies do not visibly change with time (see video corresponding to Figure 10). Since neither part of the first normal stress difference orientation appears to be time dependent, the time dependence of Figure 3 is caused by the changes in relative magnitude between the butterflies of Figure 6 and Figure 10. Figure 6 and Figure 10 are scaled differently, and specifically they happen to be scaled so as to hide these changes in relative magnitude. Reexamining the video corresponding to Figure 3, we see the orientations of both Figure 6 and Figure 10 appearing at various times during an alternant cycle, specifically at $\omega t = 2.2848, 2.9195$. We next deepen our understanding of the second normal stress difference orientation by further dissecting ψ .

C. Contribution to second normal stress difference

We next obtain the orientation distribution function components contributing to the second normal stress difference from Eq. (39) and insert them into Eq. (9), to get:

$$\psi_{N2}(\theta, \phi, t) = \frac{1}{4\pi} \left[\begin{aligned} & - (6\lambda\gamma^0)^2 \frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0, 1; t) \right] \\ & + (6\lambda\gamma^0)^4 \left[\begin{aligned} & - \frac{1}{74088} \left[\begin{aligned} & 23(0, 0, 0, 1; t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \\ & + 5 \left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t \right) + \frac{50}{11} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t \right) \end{aligned} \right] P_2^0 c_0 \\ & - \frac{1}{63504} \left[\begin{aligned} & 23(0, 0, 0, 1; t) + 12 \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \\ & + \frac{35}{3} \left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t \right) + \frac{70}{3} \left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t \right) \end{aligned} \right] P_2^2 c_2 \end{aligned} \right] \end{aligned} \right] \quad (62)$$

We call ψ_{N2} the *second normal stress difference orientation distribution*. From Eq. (62), we see that the orientations causing the second normal stress difference include only even powers of $\lambda\gamma^0$ (second and fourth order), which happens to correspond to the absence of odd powers of $\lambda\gamma^0$ in the Fourier expansion of the second normal stress difference (see Eq. (21) of [39]).

We replace the paren functions using Eqs. (41)-(54) and the Legendre polynomials using Eqs. (11)-(13) in Eq. (62) to find:

$$\psi_{N2}(\theta, \phi, t) = \frac{1}{4\pi} \left[\begin{aligned} & - \frac{1}{48} (6\lambda\gamma^0)^2 \left(\frac{1-2W}{(1+4W)(1+W)} \cos 2\omega t + \frac{3\lambda\omega \sin 2\omega t}{(1+W)} + \frac{1}{(1+W)} \right) \left(\frac{1}{7} (3C^2 - 1) c_0 - (1 - C^2) c_2 \right) \\ & + \frac{1}{241920} (6\lambda\gamma^0)^4 \left[\begin{aligned} & \left[\begin{aligned} & -50 \frac{1627128W^4 + 977292W^3 - 5781831W^2 - 4744672W + 155675}{(1+16W)(1+9W)(1+4W)(1+W)(25+9W)(100+81W)} \cos 4\omega t \\ & + 5 \frac{42485148W^3 + 99517905W^2 + 39638602W - 15077675}{(1+16W)(1+9W)(1+4W)(1+W)(25+9W)(100+81W)} \lambda\omega \sin 4\omega t \\ & + 2 \frac{27917501148W^7 + 395970769863W^6 + 1359686945763W^5 + 1401100327677W^4}{(1+4W)(1+9W)(1+W)^3(25+9W)(1+16W)(100+81W)(100+9W)} \cos 2\omega t \\ & + 4 \frac{4554095076W^7 + 54922330017W^6 + 62656044453W^5 - 366082940052W^4}{(1+4W)(1+9W)(1+W)^3(25+9W)(1+16W)(100+81W)(100+9W)} \lambda\omega \sin 2\omega t \\ & + 588 \frac{-1161W^3 - 14769W^2 - 74200W - 40000}{(1+4W)(1+W)^2(25+9W)(100+9W)} \end{aligned} \right] (3C^2 - 1) c_0 \\ & - 2 \left[\begin{aligned} & 50 \frac{29160W^4 - 11556W^3 - 107889W^2 - 63124W + 2225}{(1+16W)(1+9W)(1+4W)(1+W)(25+9W)(100+81W)} \cos 4\omega t \\ & - 5 \frac{836244W^3 + 1458315W^2 + 392806W - 211025}{(1+16W)(1+9W)(1+4W)(1+W)(25+9W)(100+81W)} \lambda\omega \sin 4\omega t \\ & - 2 \frac{506797884W^7 + 6027824709W^6 + 19477775889W^5 + 19601882811W^4}{(1+4W)(1+9W)(1+W)^3(25+9W)(1+16W)(100+81W)(100+9W)} \cos 2\omega t \\ & - 4 \frac{73929348W^7 + 725310531W^6 + 487357884W^5 - 5792477481W^4}{(1+4W)(1+9W)(1+W)^3(25+9W)(1+16W)(100+81W)(100+9W)} \lambda\omega \sin 2\omega t \\ & + 12 \frac{1107W^3 + 19303W^2 + 60050W + 28750}{(1+4W)(1+W)^2(25+9W)(100+9W)} \end{aligned} \right] (1 - C^2) c_2 \end{aligned} \right] \end{aligned} \right] \quad (63)$$

which we illustrate in Figure 5 (see (1,3), (3,1) and (3,3) of Table IV and (4,1) of Table V). As discussed in Section III.A, the lobes of Figure 5 are along the x – and y – axes and are of unequal sizes. From our video included with Figure 5, we see that the lobes along the x – axis do not evolve with time while the y – axis lobes

grow and shrink during each alternant cycle. Though the lobe sizes differ, the lobe orientations of Figure 5 match those of Figure 6, where only orientation contributing to both normal stress differences is included.

If we then isolate the parts of orientation that contribute to the second normal stress difference and not the first normal stress difference (see (3,2) of Table V, see Figure 11), we obtain a unique orientation distribution that is primarily along the z -axis. This is the only orientation distribution in this work that is principally along this axis. The orientation distributions in Figure 11 do not change throughout an alternant cycle (see our video included with Figure 11). We find that Figure 11, Figure 17 of [45] and Figure 7A of [46] nearly match. Whereas Figure 17 of [45] and Figure 7A of [46] include all orientations present in Eq. (18), Figure 11 includes orientation from both Eqs. (16) and (18). Because these figures all nearly match, we see that for the second normal stress difference the contributions from the lower orders of the orientation distribution [Eq. (16)] matter less than those of higher [Eq. (18)].

Figure 5 illustrates the evolution of the orientations contributing to the second normal stress difference. This evolution is caused by the combination of orientations common to both normal stress differences and of orientations specific to the second normal stress difference. Of course, the near absence of z -axis orientation in Figure 5 shows the greater importance of orientations contributing to both normal stresses.

D. Non-contribution to rheological measurements

In this subsection, we examine orientations that escape rheological measurement. The orientation distribution components not contributing to any rheological measurement are in (4,4) of Table IV and (4,1-3) of Table V. Noncontributing components are the parts of orientation both present in Eq. (9) and absent from Eqs. (30), (36) and (39). We thus find the noncontributing orientation distribution, ψ_{NC} , to be:

$$\psi_{NC} = \frac{1}{4\pi} \left[\begin{aligned} & (6\lambda\dot{\gamma}^0)^2 \left(\frac{1}{72} \left(\frac{6}{7} P_4^0 c_0 - \frac{1}{28} P_4^4 c_4 \right) \left(\frac{7}{3}, 1; t \right) \right) \\ & + \frac{1}{432} (6\lambda\dot{\gamma}^0)^3 \left(\begin{aligned} & - \left[\frac{3}{49} \left(\frac{7}{3}, 0, 1; t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1; t \right) \right] P_4^2 s_2 \\ & + \frac{1}{14} \left[\frac{1}{2} \left(\frac{7}{3}, 0, 1; t \right) + \frac{30}{539} \left(0, \frac{7}{3}, 1; t \right) \right] P_4^4 s_4 \\ & + \frac{1}{22} \left[P_6^2 s_2 - \frac{1}{72} P_6^6 s_6 \right] \left(\frac{11}{3}, \frac{7}{3}, 1; t \right) \end{aligned} \right) \end{aligned} \right] \quad (64)$$

We insert the paren functions from Eqs. (43)-(50) and the Legendre polynomials from Eqs. (19)-(23) to get:

$$\begin{aligned}
\psi_{\text{NC}} = & \frac{1}{4\pi} \left[(6\lambda\gamma^0)^2 \left(\frac{1}{72} \left(\frac{6}{7} P_4^0 c_0 - \frac{1}{28} P_4^4 c_4 \right) \left(\frac{3(10-6W)\cos 2\omega t + 48\lambda\omega \sin 2\omega t}{16(25+9W)(1+W)} + \frac{3}{20(1+W)} \right) \right) \right. \\
& + \frac{1}{432} (6\lambda\gamma^0)^3 \left[- \left[\frac{3}{49} \left(\frac{3(10-47W)\cos 3\omega t + 9(13-6W)\lambda\omega \sin 3\omega t}{4(1+4W)(100+81W)(1+W)} \right. \right. \right. \\
& \left. \left. + \frac{9(10+17W)\cos \omega t + 9(13+6W)\lambda\omega \sin \omega t}{4(1+4W)(100+9W)(1+W)} \right) \right] P_4^2 s_2 \\
& + \frac{1}{14} \left[\frac{1}{2} \left(\frac{3(10-47W)\cos 3\omega t + 9(13-6W)\lambda\omega \sin 3\omega t}{4(1+4W)(100+81W)(1+W)} \right. \right. \\
& \left. \left. + \frac{9(10+17W)\cos \omega t + 9(13+6W)\lambda\omega \sin \omega t}{4(1+4W)(100+9W)(1+W)} \right) \right] P_4^4 s_4 \\
& \left. + \frac{1}{22} \left[P_6^2 s_2 - \frac{1}{72} P_6^6 s_6 \right] \left(\frac{15(7-9W)\cos 3\omega t + 3(71-9W)\lambda\omega \sin 3\omega t}{8(25+9W)(49+9W)(1+W)} \right. \right. \\
& \left. \left. + \frac{3(525-19W)\cos \omega t + 3(355+3W)\lambda\omega \sin \omega t}{40(25+9W)(49+W)(1+W)} \right) \right] \Bigg] \quad (65)
\end{aligned}$$

$$\begin{aligned}
\psi_{\text{NC}} = & \frac{1}{4\pi} \left[(6\lambda\dot{\gamma}^0)^2 \left(\frac{1}{72} \left(\frac{6}{7} P_4^0 c_0 - \frac{1}{28} P_4^4 c_4 \right) \left(\frac{3(10-6W)\cos 2\omega t + 48\lambda\omega \sin 2\omega t}{16(25+9W)(1+W)} + \frac{3}{20(1+W)} \right) \right) \right. \\
& + \frac{1}{432} (6\lambda\dot{\gamma}^0)^3 \left. \left(\begin{aligned} & \frac{9}{154} \left[\begin{aligned} & \frac{1}{2} \frac{19W-5}{(25+9W)(1+W)(1+4W)} \cos 3\omega t \\ & + 3 \frac{W-3}{(25+9W)(1+W)(1+4W)} \lambda\omega \sin 3\omega t \\ & - \frac{3}{14} \frac{860+97W}{(1+4W)(100+9W)(1+W)} \cos \omega t \\ & - \frac{9}{14} \frac{256+25W}{(1+4W)(100+9W)(1+W)} \lambda\omega \sin \omega t \end{aligned} \right] P_4^2 s_2 \\ & - \frac{3}{60368} \left[\begin{aligned} & \frac{264717W^2 + 575995W - 139250}{(1+4W)(100+81W)(1+W)(25+9W)} \cos 3\omega t \\ & + 3 \frac{32346W^2 + 3597W - 178925}{(1+4W)(100+81W)(1+W)(25+9W)} \lambda\omega \sin 3\omega t \\ & - 3 \frac{9890 + 8623W}{(1+4W)(100+9W)(1+W)} \cos \omega t \\ & - 3 \frac{10757 + 3288W}{(1+4W)(100+9W)(1+W)} \lambda\omega \sin \omega t \end{aligned} \right] P_4^4 s_4 \\ & + \frac{1}{22} \left[P_6^2 s_2 - \frac{1}{72} P_6^6 s_6 \right] \left(\begin{aligned} & \frac{15(7-9W)\cos 3\omega t + 3(71-9W)\lambda\omega \sin 3\omega t}{8(25+9W)(49+9W)(1+W)} \\ & + \frac{3(525-19W)\cos \omega t + 3(355+3W)\lambda\omega \sin \omega t}{40(25+9W)(49+W)(1+W)} \end{aligned} \right) \end{aligned} \right) \right] \quad (66)
\end{aligned}$$

$$\begin{aligned}
\psi_{\text{NC}} = & \frac{1}{4\pi} \left[\frac{1}{896} (6\lambda\dot{\gamma}^0)^2 \left[\frac{(5-3W)\cos 2\omega t + 8\lambda\omega \sin 2\omega t}{2(25+9W)(1+W)} + \frac{1}{5(1+W)} \right] \left[(35C^4 - 30C^2 + 3)c_0 - 35(1-C^2)^2 c_4 \right] \right. \\
& + \frac{1}{22176} (6\lambda\dot{\gamma}^0)^3 \left. \left(\begin{aligned} & 3 \left[\begin{aligned} & \frac{1}{2} \frac{19W-5}{(25+9W)(1+W)(1+4W)} \cos 3\omega t \\ & + 3 \frac{W-3}{(25+9W)(1+W)(1+4W)} \lambda\omega \sin 3\omega t \\ & - \frac{3}{14} \frac{860+97W}{(1+4W)(100+9W)(1+W)} \cos \omega t \\ & - \frac{9}{14} \frac{256+25W}{(1+4W)(100+9W)(1+W)} \lambda\omega \sin \omega t \end{aligned} \right] P_4^2 s_2 \\ & - \frac{1}{392} \left[\begin{aligned} & \frac{264717W^2 + 575995W - 139250}{(1+4W)(100+81W)(1+W)(25+9W)} \cos 3\omega t \\ & + 3 \frac{32346W^2 + 3597W - 178925}{(1+4W)(100+81W)(1+W)(25+9W)} \lambda\omega \sin 3\omega t \\ & - 3 \frac{9890 + 8623W}{(1+4W)(100+9W)(1+W)} \cos \omega t \\ & - 3 \frac{10757 + 3288W}{(1+4W)(100+9W)(1+W)} \lambda\omega \sin \omega t \end{aligned} \right] P_4^4 s_4 \\ & + \frac{735}{64} \left[\begin{aligned} & \frac{5(7-9W)\cos 3\omega t + (71-9W)\lambda\omega \sin 3\omega t}{(25+9W)(49+9W)(1+W)} \\ & + \frac{(525-19W)\cos \omega t + (355+3W)\lambda\omega \sin \omega t}{5(25+9W)(49+W)(1+W)} \end{aligned} \right] \left[(1-C^2)(33C^4 + 18C^2 + 1)s_2 - 11(1-C^2)^3 s_6 \right] \end{aligned} \right) \right] \quad (67)
\end{aligned}$$

which is the orientation distribution function for all noncontributing components

of orientation. From Eq. (67), we create Figure 7 showing the eight-petalled flower orientation distribution, where each petal is of equal length. Although Figure 7 appears to be independent of time, the video corresponding to Figure 7 shows that the distribution does evolve through ten-lobed structures. At intermediate time $\omega t \approx 1.90$ for instance, the longest petals bisect the first and third quadrants of the xy -plane, and the shortest petals bisect the second and fourth quadrants. In the next frame, at $\omega t \approx 1.93$, the petals away from the bisection of the first and third quadrants of the xy -plane disappear, and lobes outside the xy -plane appear. These new lobes are smaller than the two major petals bisecting the xy -plane. At frame $\omega t \approx 1.96$, the petal lengths invert so that the longest petals are now bisecting the second and fourth quadrants of the xy -plane. To fully appreciate this sequence of orientations that escape rheological measurement, one really must view carefully the video included with Figure 7. Look for this sequence of orientations occurring over $1.90 \leq \omega t \leq 1.96$ to repeat itself over $5.03 \leq \omega t \leq 5.09$.

We discover a second departure from the eight-petalled flower orientation near $\omega t \approx 2.53$ and 5.68 . Here, the same sequence of reorientations is reversed from that of the previous paragraph. These sequences of molecular orientations escape rheological measurement (τ_{yx} , N_1 and N_2) and this is why emerging scattering measurements in LAOS are so important [27].

From Eq. (67), in the limit as $\lambda\dot{\gamma}^0 \rightarrow 0$, we learn that the noncontributing part of the orientation distribution disappears. In other words, when we measure $\eta^*(\omega)$, all of the molecules contribute. Further, the larger the $\dot{\gamma}^0$ of the oscillatory shear flow, the greater the fraction of polymer that escapes our rheological measurement.

IV. CONCLUSION

We have deepened our understanding of the molecular motion underlying the rheological measurements associated with large-amplitude oscillatory shear flow, specifically, the shear stress and normal stress differences. We have explored each part of orientation and how each of these parts contributes to each rheological response or not. We did this for a dilute suspension of rigid dumbbells in a Newtonian fluid, the simplest relevant model. We developed three-dimensional orientation distributions to explore the contribution (Figure 3 - Figure 5) and non-contribution (Figure 7) of polymer orientation to rheological measurements.

We find that some polymer orientations escape rheological measurements. By *escape*, we mean that the noncontributing orientation distribution contains orientations that contribute to neither the shear stress nor the normal stress differences. Through our 3D imagery and included videography (Figure 7), we uncovered the eight-petalled shape of these rheological escapees.

Orientations contributing to the shear stress resemble those of the total orientation distribution, and all orientations featured in both normal stress difference orientation distributions are included in the shear stress. In other

words, the normal stress differences provide information about specific parts of the shear stress orientation distribution. We further dissected these parts of orientation by way of Table IV and Table V. Each of these dissections perfects our understanding of how molecular orientation causes fluid elasticity in LAOS.

We have limited the scope of this work to the alternant part of the orientation distribution function in LAOS. It would of course be interesting to explore how the fluid orientation progress from quiescence to alternance. We leave this study of orientation in LAOS start-up for another day.

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Table I: Summary of orientation contribution and non-contribution to shear stresses and normal stress differences in the literature

Harmonic	Model	Expansion Variable	Orientation Harmonic	Non-contributing terms	Fourier Decomposition	Shear Stress	Normal Stress Differences	[Ref.] (Correction to)
Kirkwood and Plock (1956, 1967); Plock (1957)	RD^h, SK^h	$\dot{\gamma}^0$	0,2,4,6			X		[28,29,30]
Paul (1969); Paul (1970); Bharadwaj (2012)	RD^h, SK^h	$\dot{\gamma}^0$	0,2,4,6			X		[31,32,33] (28,30)
Paul and Mazo (1969), Paul (1970)	RR	$\dot{\gamma}^0$	0,2,4,6			X		[34,32]
Bird, Warner and Evans (1971)	RD, SK	$\dot{\gamma}^0$	0,1,2,3			X	X	[25]
Mou and Mazo (1977)	RR	$\dot{\gamma}^0$	0,2,4,6				X	[35](34)
Helfand and Rochefort (1982)	$R^\square$	$\dot{\gamma}^0$	See Eq. (8)			X		[36]
Bird <i>et al.</i> (2014)	RD, SK	$\dot{\gamma}^0$	0,1,2,3			X		[37]
Schmalzer <i>et al.</i> (2014, 2015)	RD, SK	$\dot{\gamma}^0$	0,1,2,3,4				X	[38,39]
Schmalzer <i>et al.</i> (2014, 2015)	RD, SK	$\dot{\gamma}^0$	0,1,2,3,4	X				[40,41,42]
Bozorgi (2014); Bozorgi and Underhill (2014)	AS^h	γ_0	0,1,2			X		Chapter 8 of [43]; [44]
Giacomin <i>et al.</i> (2015)	RD, SK	$\dot{\gamma}^0$	0,1,2,3,4	X	X			[45]
Gilbert and Giacomin (2016)	RD, SK	$\dot{\gamma}^0$	$0,1',1'',2',2'',3',3'',4',4''$	X	X			[46]
Jbara <i>et al.</i> (2016)	RD, SK	$\dot{\gamma}^0$	0,1,2,3,4		X	X		[47]
This Paper	RD, SK	$\dot{\gamma}^0$	0,1,2,3,4	X		X	X	

Legend: AS = active rod suspensions; R = reptation; RD = rigid dumbbell; RR = planar rigid ring; SK = shish-kebab; $\square$ = mixtures; ^{h} = with hydrodynamic interaction; n' = in-phase with $\cos n\omega t$; n'' = out-of-phase with $\cos n\omega t$.

Table II: Dimensional Variables

Velocity, i th component	L/t	v_i
Angular frequency	t^{-1}	ω
Bead center to center length of rigid dumbbell	L	L
Bead diameter	L	b
Bead friction coefficient	M/t	$\zeta \equiv 3\pi b\eta_s$
Boltzmann constant	$ML^2/t^2T/\text{dumbbell}$	k
Cartesian coordinate, distance from stationary plate (Figure 2)	L	y
Cartesian coordinate, flow direction (Figure 2)	L	x
Cartesian coordinate, transverse to flow direction (Figure 2)	L	z
Gap	L	h
Relaxation time of fluid	t	λ
Shear rate amplitude	t^{-1}	$\dot{\gamma}^0$
Solvent viscosity	M/Lt	η_s
Temperature	T	T
Extra stress component ^a	M/Lt^2	τ_{ij}
Shear stress, reference	M/Lt^2	$\tau_{yx,s}$
Time	t	t

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

^aWhere τ_{ij} is the force exerted in the j th direction on a unit area of a fluid surface of constant x_i from fluid of lower x_i on fluid of greater x_i [48].

Table III: Dimensionless Variables and Groups

Azimuthal angle	ϕ
Deborah number squared	$W \equiv (\lambda\omega)^2$
Deborah number, oscillatory shear	$\lambda\omega$
Number of equally-spaced beads on a rod of length $(L - b)$	N
Number of harmonic	n
Orientation distribution function	$\psi(\theta, \phi, t)$
Shear stress orientation distribution function	$\psi_{ss}(\theta, \phi, t)$
First normal stress difference orientation distribution function	$\psi_{N1}(\theta, \phi, t)$
Second normal stress difference orientation distribution function	$\psi_{N2}(\theta, \phi, t)$
Noncontributing orientation distribution function	$\psi_{NC}(\theta, \phi, t)$
i th term of orientation distribution expansion	$\psi_p(\theta, \phi, t)$
Orientation distribution, Fourier component, in-phase with $\cos n\omega t$	$\psi'_n(\theta, \phi)$
Orientation distribution, Fourier component, out-of-phase with $\cos n\omega t$	$\psi''_n(\theta, \phi)$
Shear strain amplitude	γ_0
Polar angle	θ
Legendre polynomial [Eqs. (11)-(13) and Eqs. (19)-(23)]	P_n^m
Weissenberg number, oscillatory shear	$\lambda\dot{\gamma}^0$

Table IV: Summary of orientation contribution and non-contribution to shear stresses and normal stress differences, where each entry represents the spherical harmonic present in both the **first column** and **top row**

	τ_{yx} Eq. (30)	$\tau_{xx} - \tau_{yy}$ Eq. (36)	$\tau_{yy} - \tau_{zz}$ Eq. (39)	[-]
τ_{yx} Eq. (30)	$P_0^0 c_0$ $P_2^0 c_0$ $P_2^2 s_2$ $P_2^2 c_2$ Figure 3	$P_2^2 s_2$ $P_2^2 c_2$ Figure 4	$P_2^0 c_0$ $P_2^2 c_2$ Figure 5	X
$\tau_{xx} - \tau_{yy}$ Eq. (36)	$P_2^2 s_2$ $P_2^2 c_2$ Figure 4	$P_2^2 s_2$ $P_2^2 c_2$ Figure 4	$P_2^2 c_2$ Figure 6	X
$\tau_{yy} - \tau_{zz}$ Eq. (39)	$P_2^0 c_0$ $P_2^2 c_2$ Figure 5	$P_2^2 c_2$ Figure 6	$P_2^0 c_0$ $P_2^2 c_2$ Figure 5	X
[-]	X	X	X	$P_4^0 c_0$ $P_4^2 s_2$ $P_4^4 s_4$ $P_4^4 c_4$ $P_6^2 s_2$ $P_6^6 s_6$ Figure 7

Legend:

Table V: Summary of orientation contribution and non-contribution to shear stresses and normal stress differences, where each entry represents the spherical harmonic present in the **first column** but absent from the **top row**

	τ_{yx} Eq. (30)	$\tau_{xx} - \tau_{yy}$ Eq. (36)	$\tau_{yy} - \tau_{zz}$ Eq. (39)	[-]
τ_{yx} Eq. (30)	X	$P_0^0 c_0$ $P_2^0 c_0$ Figure 8	$P_0^0 c_0$ $P_2^2 s_2$ Figure 9	$P_0^0 c_0$ $P_2^0 c_0$ $P_2^2 s_2$ $P_2^2 c_2$ Figure 3
$\tau_{xx} - \tau_{yy}$ Eq. (36)	X	X	$P_2^2 s_2$ Figure 10	$P_2^2 s_2$ $P_2^2 c_2$ Figure 4
$\tau_{yy} - \tau_{zz}$ Eq. (39)	X	$P_2^0 c_0$ Figure 11	X	$P_2^0 c_0$ $P_2^2 c_2$ Figure 5
[-]	$P_4^0 c_0$ $P_4^2 s_2$ $P_4^4 s_4$ $P_4^4 c_4$ $P_6^2 s_2$ $P_6^6 s_6$ Figure 7	$P_4^0 c_0$ $P_4^2 s_2$ $P_4^4 s_4$ $P_4^4 c_4$ $P_6^2 s_2$ $P_6^6 s_6$ Figure 7	$P_4^0 c_0$ $P_4^2 s_2$ $P_4^4 s_4$ $P_4^4 c_4$ $P_6^2 s_2$ $P_6^6 s_6$ Figure 7	X

Legend:

Figure 1: The dumbbell consists of identical beads (of identical mass and of identical diameter) connected by one thin, absolutely rigid (dimensionless, massless) rod.

Figure 2: Orthomorphic sketch of alternating velocity profile in oscillatory shear between stationary and moving plates [Eqs. (2) or (3)]. Cartesian coordinates with origin on the stationary plate.

Figure 3: Orientation distribution for components contributing to the shear stress, ψ_{ss} , from Eq. (40), (1,1) of **Table IV** and (1,4) of **Table V**, over a complete alternant cycle, $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 1$, where the first five orientation expansion terms contribute.

Figure 4: Orientation distribution for components contributing to both the shear stress, ψ_{ss} , and the first normal stress difference, ψ_{N1} , from Eq. (61), (1,2), (2,1), and (2,2) of **Table IV** and (2,4) of **Table V** over a complete alternant cycle, $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 1$, where four orientation expansion terms contribute (second through fifth).

Figure 5: Orientation distribution for components contributing to the shear stress, ψ_{ss} , and the second normal stress difference, ψ_{N_2} , from Eq. (63), (1,3), (3,1), and (3,3) of **Table IV** and (3,4) of **Table V**, over a complete alternant cycle, $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 1$, where the third and fifth orientation expansion terms contribute.

Figure 6: Orientation distribution for components contributing to both the first, ψ_{N1} , and second, ψ_{N2} , normal stress differences from (2,3) and (3,2) of **Table IV** and (2,4), over a complete alternant cycle, $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 1$, where only the fifth orientation expansion term contributes.

Figure 7: Orientation distribution for components not contributing to any rheological measurement from Eq. (67), (4,4) of **Table IV** and (4,1-3) of **Table V**, over a complete alternant cycle, $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 1$, where the third and fourth orientation expansion terms contribute.

Figure 8: Orientation distribution for components contributing to the shear stress, ψ_{ss} , but not to the first normal stress difference, ψ_{N1} , from (1,2) of **Table V**, over a complete alternant cycle, $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 1$, where the first, third and fifth orientation expansion terms contribute.

Figure 9: Orientation distribution for components contributing to the shear stress, ψ_{ss} , but not to the second normal stress difference, ψ_{N_2} , from (1,3) of **Table V**, over a complete alternant cycle, $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\gamma^0 = 1$, where the first, second, and fourth orientation expansion terms contribute.

Figure 10: Orientation distribution for components contributing to the shear stress, ψ_{N1} , but not to the second normal stress difference, ψ_{N2} , from (2,3) of **Table V**, over a complete alternant cycle, $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\gamma^0 = 1$, where the second and fourth orientation expansion terms contribute.

Figure 11: Orientation distribution for components contributing to the second normal stress difference, ψ_{N_2} , but not to the first normal stress difference, ψ_{N_1} , from (3,2) of **Table V**, over a complete alternant cycle, $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 1$, where the first, third and fifth orientation expansion terms contribute.

VI. REFERENCES

- ¹ Gemant, A., "Komplexe Viskosität," *Naturwissenschaften* **25**, 406-407 (1935).
- ² Gemant, A., "The conception of a complex viscosity and its application to dielectrics," *Transactions of the Faraday Society*, No. 175, **XXXI**, Part II, 1582-1590 (November, 1935b); Erratum: The footnote on p. 1583, "¹⁵A. Gemant, *Naturwiss*, 1935, **23**, 406." should be "¹⁵A. Gemant, *Naturwiss.*, 1935, **25**, 406."
- ³ Bird, R.B. and A.J. Giacomin, "Who Conceived the Complex Viscosity?," *Rheologica Acta*, **51**(6), 481-486 (2012).
- ⁴ Ferry, J.D. (1961) *Viscoelastic properties of polymers*, 1st edn. John Wiley & Sons, Inc., New York
- ⁵ Dealy, J.M. (1982) *Rheometers for Molten Plastics: A Practical Guide to Testing and Property Measurement*, Van Nostrand, New York.
- ⁶ Dealy, J.M and J. Wang, *Melt Rheology and its Applications in the Plastics Industry*, 2nd ed., Springer, Dordrecht (2013).
- ⁷ Wagner, M.H., V.H. Rolón-Garrido, K. Hyun, and M. Wilhelm, "Analysis of medium amplitude oscillatory shear data of entangled linear and model comb polymers," *J. Rheol.*, **55**(3), 495-516 (2011).
- ⁸ Schrag, J.L. "Measurement of the oscillatory shear birefringence of polystyrene solutions," PhD Thesis, Oklahoma State University, Stillwater, OK (May, 1967).
- ⁹ Weissenberg, K., "Rheology of Hydrocarbon Gels," *Proceedings of the Royal Society of London. Series A, Mathematical and Physical Sciences*, **200**(1061) 183-188 (Jan. 6, 1950).
- ¹⁰ Weissenberg, K., Proc., 1st Int. Congr. Rheol., Holland 1-29 (1948/9).
- ¹¹ Hyun, K., M. Wilhelm, C. O. Klein, K. S. Cho, J. G. Nam, K. H. Ahn, S. J. Lee, R. H. Ewoldt, and G. H. McKinley, "A review of nonlinear oscillatory shear tests: Analysis and application of large amplitude oscillatory shear (LAOS)," *Progress in Polymer Science*, **36**, 1697-1753 (2011); doi: 10.1016/j.progpolymsci.2011.02.002
- ¹² Giacomin, A.J., T. Samurkas, and J.M. Dealy, "A Novel Sliding Plate Rheometer for Molten Plastics," *Polymer Engineering and Science*, **29**(8), 499-504 (April, 1989).
- ¹³ Giacomin, A.J., "A Sliding Plate Melt Rheometer Incorporating a Shear Stress Transducer," PhD Thesis, Dept. Chemical Engineering, McGill University, Montréal, Canada (1987).

¹⁴ Giacomini, A.J. and J.M. Dealy, "A New Rheometer for Molten Plastics," S.P.E. Tech. Papers, XXXII, Proc. 44th Annual Tech. Conf., Society of Plastics Engineers, Boston, MA (May, 1986), pp. 711-714.

¹⁵ Roberts, J.E., "The Early Development of the Rheogoniometer," in Harris, J., ed., *Karl Weissenberg: 80th Birthday Celebration Essays*, East African Literature Bureau, Kampala (1973), pp. 153-163.

¹⁶ Park, O.O. and G.G. Fuller, "Dynamics of Rigid Dumbbells in Confined Geometries: Part II. Time-Dependent Shear Flow," *J. Non-Newton. Fluid*, **18** 111 (1985).

¹⁷ Park, O.O., "Dynamics of Rigid and Flexible Polymer Chains: Part 1. Transport Through Confined Geometries," PhD Thesis, Chemical Engineering, Stanford University, Stanford, CA (June, 1985).

¹⁸ Giacomini, A.J. and J.M. Dealy, "Using Large-Amplitude Oscillatory Shear," Chapter 11, Collyer, A.A. and D.W. Clegg, eds., *Rheological Measurement*, 2nd ed., Kluwer Academic Publishers, Dordrecht, Netherlands, pp. 327-356 (1998).

¹⁹ Giacomini, A.J. and J.M. Dealy, "Large-Amplitude Oscillatory Shear," Chapter 4, Collyer, A.A., ed., *Techniques in Rheological Measurement*, Chapman and Hall, London & New York, pp. 99-121 (1993); Kluwer Academic Publishers, Dordrecht, pp. 99-121 (1993); Erratum: Corrections to Figs 11.5-11.7 are in [18] above.

²⁰ Ewoldt, R.H., "Defining nonlinear rheological material functions for oscillatory shear," *Journal of Rheology*, **57**, 177-195 (2013).

²¹ Giacomini, A.J., R.B. Bird, L.M. Johnson and A.W. Mix, "Large-Amplitude Oscillatory Shear Flow from the Corotational Maxwell Model," *Journal of Non-Newtonian Fluid Mechanics*, **166**(19-20), 1081-1099 (2011). Errata: after Eq. (20), Ref. [10] should be [13]; 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 (97) above replace Figs. 5-7; on the ordinates of Figs. 5-7, $\frac{1}{2}$ should be 2; after Eq. (119), " $(\xi\alpha)$ " should be " $\xi(\alpha)$ "; in Eq. (147), " $n - 1$ " should be " $n = 1$ "; in Eqs. (76) and (77), Ψ' and Ψ'' should be Ψ_1' and Ψ_1'' ; throughout, Ψ_1^d , Ψ_1' and Ψ_1'' should be Ψ_1^d , Ψ_1' and Ψ_1'' ; in Eq. (127), " $\cos\tau$ " should be " $(-\cos\tau)$ "; 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 Eqs. (181) and (182), "1,21"

should be “1,2”; in Eqs. (184) and (185), “ mp ” should be “1, mp ”; Eq. (65) should be $We/De > \sqrt[n_N]{(h_N + 1)!}$; see also [22] below.

²² Giacomini, A.J., R.B. Bird, L.M. Johnson and A.W. Mix, ‘Corrigenda: “Large-Amplitude Oscillatory Shear Flow from the Corotational Maxwell Model,” [Journal of Non-Newtonian Fluid Mechanics, **166**, 1081–1099 (2011)],’ *Journal of Non-Newtonian Fluid Mechanics*, **187-188**, 48-48 (2012); see also [21] above.

²³ Bird, R.B. and R.C. Armstrong, “Time-Dependent Flows of Dilute Solutions of Rodlike Macromolecules,” *The Journal of Chemical Physics*, **56**, 3680 (1972).

Addendum: In Eq. (8), the term $-\frac{1}{432}\left[\frac{3}{49}\left(\frac{7}{3}, 0, 1; t\right) + \frac{30}{539}\left(0, \frac{7}{3}, 1; t\right)\right]P_4^2s_2$ should be inserted just before the “+ additional terms”.

²⁴ Bird, R.B., O. Hassager, R.C. Armstrong, and C.F. Curtiss, *Dynamics of polymeric liquids*, Vol. 2, 1st ed., Wiley, New York (1977). Erratum: In Problem **11.C.1 d.**, “and ϕ_2 ” should be “through ϕ_4 ”; On p. N-2, under L entry “Length of a rigid dumbbell connector” should be “Bead center to center length of a rigid dumbbell”.

²⁵ Bird, R.B., H.R. Warner, Jr., and D.C. Evans, “Kinetic theory and rheology of dumbbell suspensions with Brownian motion,” *Advances in Polymer Science* (or *Fortschr. Hochpolymeren-Forschung.*), **8**, 1 (1971).

²⁶ Bird, R.B., C.F. Curtiss, R.C. Armstrong, and O. Hassager, *Dynamics of polymeric liquids*, vol 2, 2nd edn., John Wiley & Sons, Inc., New York (1987). Erratum: On p. 409 of the first printing, the $(n + m)!$ in the denominator should be $(n - m)!$; In TABLE 16.4-1, under L entry “length of rod” should be “bead center to center length of a rigid dumbbell”; In the Figure 14.1-2 caption, “Multibead rods of length L ” should be “Multibead rods of length $L + d$ ”.

²⁷ Lettinga, M.P., P. Holmqvist, P. Ballesta, S. Rogers, D. Kleshchanok, and B. Struth, “Nonlinear behavior of nematic platelet dispersions in shear flow,” *Physical Review Letters*, **109**(24), 246001 (2012).

²⁸ Kirkwood, J.G. and R.J. Plock, “Non-Newtonian viscoelastic properties of rod-like macromolecules in solution,” *J. Chem. Phys.*, **24**, 665-669 (1956).

²⁹ Auer, P.L. (Ed.), *Macromolecules (John Gamble Kirkwood Collected Works)*, J. G. Kirkwood and R. J. Plock, “Non-Newtonian viscoelastic properties of rod-like macromolecules in solution,” Gordon and Breach, New York (1967). Errata: On the left side of Eq. (1) on p. 113, ϵ should be $\dot{\epsilon}$. See also Eq. (1) of [Error! Bookmark not defined.]; In Eq. (2a), G' should be G'' , and in Eq. (2b), G'' should be G' . See Eqs. (117a) and (117b) of [Error! Bookmark not defined.].

³⁰ R.J. Plock, "I. Non-Newtonian Viscoelastic Properties of Rod-Like Macromolecules in Solution. II. The Debye-Hückel, Fermi-Thomas Theory of Plasmas and Liquid Metals," PhD Thesis, Yale University, New Haven, CT (June, 1957). Errata: In Eqs. (2.4a), G' should be G'' , and in Eq. (2.4b), G'' should be G' . See Eqs. (117a) and (117b) of [Error! Bookmark not defined.].

³¹ E. Paul, "Non-Newtonian viscoelastic properties of rodlike molecules in solution: comment on a paper by Kirkwood and Plock," *J. Chem. Phys.*, **51**, 1271-1290 (1969).

³² E.W. Paul, "Some Non-Equilibrium Problems for Dilute Solutions of Macromolecules; Part I: The Plane Polygonal Polymer," PhD Thesis, Dept. of Chemistry, University of Oregon, Eugene, OR (September, 1970).

³³ N.A.K. Bharadwaj, "Low Dimensional Intrinsic Material Functions Uniquely Identify Rheological Constitutive Models And Infer Material Microstructure," Masters Thesis, Mechanical Engineering, University of Illinois at Urbana-Champaign, IL (2012).

³⁴ E.W. Paul and R.M. Mazo, "Hydrodynamic Properties of a Plane-Polygonal Polymer, According to Kirkwood-Riseman Theory," *J. Chem. Phys.*, **51**, 1102 (1969).

³⁵ C.Y. Mou and R.M. Mazo, "Normal stress in a solution of a plane-polygonal polymer under oscillating shearing flow," *J. Chem. Phys.*, **67**, 5972 (1977).

³⁶ E. Helfand and D.S. Pearson, "Calculation of the nonlinear stress of polymers in oscillatory shear fields," *J. Polym. Sci.: Pol. Phys. Ed.*, **20**, 1249-1258 (1982).

³⁷ R.B. Bird, A.J. Giacomin, A.M. Schmalzer and C. Aumnate, "Dilute Rigid Dumbbell Suspensions in Large-Amplitude Oscillatory Shear Flow: Shear Stress Response," *The Journal of Chemical Physics*, **140**, 074904 (2014); Corrigenda: In Eq. (91), η' should be η'' ; In caption to Fig. 3, " $\psi_1[P_2^2 s_2]$ " should be " $\cos 3\omega t$ " and " $\psi_2[P_2^0 c_0, P_2^2 c_2, \dots]$ " should be " $\sin 3\omega t$ ".

³⁸ A.M. Schmalzer, R.B. Bird and A.J. Giacomin, "Normal Stress Differences in Large-Amplitude Oscillatory Shear Flow for Dilute Rigid Dumbbell Suspensions," PRG Report No. 002, QU-CHEE-PRG-TR--2014-2, Polymers Research Group, Chemical Engineering Dept., Queen's University, Kingston, CANADA (April, 2014).

³⁹ Schmalzer, A.M., R.B. Bird and A.J. Giacomin, "Normal Stress Differences in Large-Amplitude Oscillatory Shear Flow for Dilute Rigid Dumbbell Suspensions,"

Journal of Non-Newtonian Fluid Mechanics, **222**, 56-71 (2015); Errata: Above Eqs. (14) and (25), “significant figures” should be “16 significant figures”.

⁴⁰ A.M. Schmalzer and A.J. Giacomin, “Orientation in Large-Amplitude Oscillatory Shear,” *Macromolecular Theory and Simulations*, **24**(3), 171, 181-207 (May, 2015). Errata: From 6th line of Introduction, delete “small-amplitude oscillatory shear”; in Ref. [22], “[23]” should be “[21]”

⁴¹ A.M. Schmalzer and A.J. Giacomin, “Orientation in Large-Amplitude Oscillatory Shear,” PRG Report No. 005, QU-CHEE-PRG-TR--2014-5, Polymers Research Group, Chemical Engineering Dept., Queen’s University, Kingston, CANADA (June, 2014).

⁴² A.M. Schmalzer, “Large-Amplitude Oscillatory Shear Flow of Rigid Dumbbell Suspensions,” PhD Thesis, University of Wisconsin, Mechanical Engineering Dept., Madison, WI (August 8, 2014).

⁴³ Y. Bozorgi, “Multiscale Simulation of the Collective Behavior of Rodlike Self-Propelled Particles in Viscoelastic Fluids,” PhD Thesis, Chemical and Biological Engineering, Rensselaer Polytechnic Institute, Troy, NY (2014).

⁴⁴ Y. Bozorgi and P.T. Underhill, “Large-amplitude oscillatory shear rheology of dilute active suspensions,” *Rheologica Acta*, DOI 10.1007/s00397-014-0806-y (2014).

⁴⁵ A.J. Giacomin, P.H. Gilbert, and A.M. Schmalzer, “Fourier decomposition of polymer orientation in large-amplitude oscillatory shear flow,” *Structural Dynamics*, **2**, 024101 (2015), pp. 1-20.

⁴⁶ Gilbert, P.H. and A.J. Giacomin, “Complex Polymer Orientation,” *Polymer*, doi:10.1016/j.polymer.2016.05.046 (May 29, 2016).

⁴⁷ Jbara, L., P.H. Gilbert, and A.J. Giacomin, “The fifth shear-stress harmonic for polymeric liquids in large-amplitude oscillatory shear flow,” *Nihon Reoroji Gakkaishi*, **44**(5), xx-xx (2016). In Press.

⁴⁸ Bird, R.B., W.E. Stewart, and E.N. Lightfoot, *Transport phenomena*, Revised 2nd ed., Wiley and Sons, New York (2007).