QUEEN'S UNIVERSITY
POLYMERS RESEARCH GROUP
19 Division Street, Kingston, ON, K7L 3N6 Canada

ORIENTATION IN LARGE-AMPLITUDE OSCILLATORY SHEAR

A.M. Schmalzer^{1,2} and A.J. Giacomin^{3,4,*}

¹Rheology Research Center and ²Mechanical Engineering Department
University of Wisconsin-Madison, Madison, WI 53706

³Chemical Engineering Department and ⁴Polymers Research Group
Queen's University, Kingston, ON K7L 3N6

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*Corresponding author (giacomin@queensu.ca)

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¹Rheology Research Center

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ABSTRACT

We examine the simplest relevant molecular model for large-amplitude oscillatory shear flow of a polymeric liquid: the dilute suspension of rigid dumbbells in a Newtonian solvent. We find explicit analytical expressions for the orientation distribution function, and specifically for test conditions of frequency and shear rate amplitude that generate higher harmonics in the shear stress and normal stress difference responses. Our analysis employs the general method of Bird and Armstrong (1972) for analyzing the orientation of the rigid dumbbells in suspension in any unsteady shear flow. We have solved the diffusion equation analytically to explore the orientations of the molecules induced by the oscillatory shear flow.

We see that the orientation distribution function is neither even nor odd. We find zeroth, first, second, third and fourth harmonics of the orientation distribution function, and we have derived explicit analytical expressions for these. We use our analytical solution to examine the detailed shape of the orientation distribution function. We provide a clear visualization of the orientation distribution function in large-amplitude oscillatory shear flow in spherical coordinates all the way around one full alternant cycle (at $\omega t = 0, \frac{\pi}{4}, \frac{\pi}{2}, \dots, 2\pi$). Our analysis supplements our previous results for the shear stress [Bird et al., *JCP*, 140, 074904 (2014)] and normal stress differences [Schmalzer *et al.*, PRG Report No. 002, Queen's University (April 2014)] for a suspension of rigid dumbbells in large-amplitude oscillatory shear flow. This exploration includes the Newtonian, the linear viscoelastic and nonlinear viscoelastic regimes. We find the orientation distribution for Newtonian behavior to be nearly isotropic (spherical), for the linearly viscoelastic behavior, only slightly anisotropic (only a slight ellipsoidal departure from spherical), and for the nonlinear viscoelastic regime, we find the orientation distribution function for the dumbbells to be highly anisotropic (and even peanut or dumbbell shaped, which we call lemniscoidal).

*Corresponding author (giacomin@queensu.ca).

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I. INTRODUCTION

Since its conception in the 1930s [1,2,3], and then the perfection of the measurement [4], oscillatory shear flow testing became, by far, the most common viscoelastic property measurement [5,6,7]. A great deal is understood about the structure-property relations for polymer molecules in small-amplitude oscillatory shear small-amplitude oscillatory shear flow [8,9]. Comparatively little is known about the behavior of the molecules in large-amplitude oscillatory shear flow. Our previous work on large-amplitude oscillatory shear derives expressions for the shear stress and for both normal stress difference responses, using both continuum [10,11] and molecular approaches [12,13,14]. For the molecular approach, we use the simplest relevant model of a polymeric liquid, the dilute suspension of rigid dumbbells. By *simplest relevant*, we mean the simplest model that gives the appropriate higher harmonics in the shear stress and normal stress differences [12]. We choose rigid dumbbells over Hookean dumbbells, because Hookean dumbbells predict no higher harmonics in large-amplitude oscillatory shear at all.

A rigid dumbbell has both length and width, the two key geometric properties of a polymer molecule, and these properties confer upon the molecule orientation. In our previous work, we have used the diffusion equation for molecular orientation to derive expressions for the shear stress and for both normal stress difference responses (see Eq. (82) of [12] and Eqs. (13) and (21) of [13]). In this paper, we will examine the orientation distribution directly, by deriving expressions for the zeroth, first, second, third and fourth harmonics of the orientation in large-amplitude oscillatory shear flow, as functions of the dimensionless frequency, $\lambda\omega$, and dimensionless shear rate amplitude, $\lambda\dot{\gamma}^0$. Our analytical solution, presented with three dimensional color graphics, gives us insight into the molecular behaviors underpinning the rheological responses [15]. We thus derive new structure-property relations for the orientation distribution in large-amplitude oscillatory shear flow.

The rigid dumbbell suspension seems to mimic the behavior of polymer solutions and melts, at least qualitatively, and this seems to be because the overall orientation of the polymer chains gives the longest time constant for the polymeric liquids.

This paper concerns itself with the orientation distribution function of a rigid dumbbell, when the flow is given by a velocity field $v_x(y, t)$. We further restrict this paper to the periodic response, reached several cycles after startup of oscillatory shear, called alternance (see Section 6. of [10]). This work is an extension of recent articles [12, 13] wherein the expressions for the shear stress response is derived using the method of Bird and Armstrong [16] (see also Problem 11C.1 of [17]).

a. The flow field

We are concerned here with a particular case of a homogeneous shearing flow (see Figure 1):

$$v_x = \dot{\gamma}^0 f(t) y, v_y = v_z = 0 \quad (1)$$

called simple shear flow. The symbols used here and elsewhere are defined, along with their dimensions, in Table I and Table II. The coefficient of proportionality between the velocity and position is $\dot{\gamma}^0 f(t)$, where the special variable f depends solely on the time. This situation can be obtained by having the flow take place between two plates placed extremely close to one another [18,19].

We consider only the case where $f(t) = \cos \omega t$ so that Eq. (1) becomes [20,21]:

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

which we call oscillatory shear flow. That is, the shear rate may be written as:

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

in which the dimensionless shear rate $\lambda \dot{\gamma}^0$ (the Weissenberg number) and the dimensionless frequency $\lambda \omega$ (the Deborah number), appear [20,21,22]. All orientation distribution responses calculated herein are thus referred to this cosinusoidal shear rate (see Section V of [23]). These dimensionless groups contain the characteristic time constant λ which, for the dilute suspension of rigid dumbbells in a Newtonian solvent of Newtonian viscosity η_s is:

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

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 beads on the rod of length L , T is absolute temperature, and 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 2). Eq. (3) applies for continuum, and thus cannot apply when the gap, h , approaches the size of the molecules, L , or when $L/h \ll 1$. For a detailed study of small-amplitude oscillatory shear for a rigid dumbbell suspension in confined gaps, see Section 5. of [24] or Section 2.3.3 of [25].

In this paper, we consider the special case of Eq. (3) where higher harmonics arise in both the shear stress and in the normal stress difference responses.

II. ORIENTATION DISTRIBUTION FUNCTION FOR UNSTEADY SHEAR FLOWS

For simple shear flow, the *diffusion equation* for the orientation distribution function $\psi(\theta, \phi, t)$ is given by (see Eq. 5.1 of [26], Eq. (14.4-1) of [27]):

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

$$\equiv \Lambda \psi - 6\lambda \dot{\gamma} \Omega \psi$$

where $\lambda \dot{\gamma}$ is a dimensionless shear rate [see Eq. (3)]. In Eq. (6), Λ and Ω are linear operators; we use the abbreviations $S \equiv \sin \theta$, $C \equiv \cos \theta$, $s \equiv \sin \phi$ and $c \equiv \cos \phi$. The dumbbell orientation is given in terms of the spherical coordinates θ and ϕ which are defined in Figure 2. When the fluid is at rest, all orientations are equally likely, and ψ is equal to a normalization constant, $1/4\pi$, so that:

$$\int_0^{2\pi} \int_0^\pi \psi(\theta, \phi, t) \sin \theta d\theta d\phi = 1 \quad (7)$$

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

To our knowledge, Eq. (6) has not been solved numerically, not for large-amplitude oscillatory shear flow, not even for steady shear flow. In this paper, we thus develop the analytical solution to Eq. (6) for large-amplitude oscillatory shear flow, using the method of Bird and Armstrong [16].

Note on notation: For ψ we adhere to the definition of Bird-Warner-Evans [26] which differs from that of [17,27]. The relation between the distribution function used here, ψ , and the symbol used in the first [17] and second [27] editions of *Dynamics of Polymeric Liquids* is (see footnote 1 on p. 524 of [17]):

$$\psi_{\text{DPL},1e} = \psi_{\text{DPL},2e} = \psi \sin \theta \quad (8)$$

Also, whereas the symbol for the distribution function used here is ψ , that used in the second edition of *Dynamics of Polymeric Liquids* [27] is f .

III. SPHERICAL HARMONICS, Λ – OPERATORS AND Ω – OPERATORS

We will find it useful to write the orientation distribution function in terms of spherical harmonics, $P_n^m c_m$ and $P_n^m s_m$, where $c_m \equiv \cos m\phi$ and $s_m \equiv \sin m\phi$, and where the P_n^m are the associated Legendre polynomials, which are functions of $C \equiv \cos \theta$ (see Table III) and which are generated by (see Chapter 8 and 22 of [28]):

$$P_n^m(C) \equiv \frac{(-1)^m}{2^n n!} (1-C^2)^{m/2} \frac{d^{n+m}}{dC^{n+m}} (C^2-1)^n \quad (9)$$

where the sign convention is dictated by the factor $(-1)^m$, called the Condon-Shortley phase [29]. The spherical harmonics obey the following orthogonality relations:

$$\int_0^{2\pi} \int_0^\pi P_n^m \begin{Bmatrix} c_m \\ s_m \end{Bmatrix} P_{n'}^{m'} \begin{Bmatrix} c_{m'} \\ s_{m'} \end{Bmatrix} \sin \theta d\theta d\phi = \frac{2\pi(n+m)!}{(2n+1)(n-m)!} \delta_{nn'} \delta_{mm'} (1 \pm \delta_{m0}) \quad (10)$$

where the positive sign is used if c 's appear in the integral, and the negative sign is used if s 's appear in the integral. The value of the integral is zero if the two spherical harmonics differ [27].

The Λ - operator acting on a spherical harmonic gives the same spherical harmonic multiplied by $-n(n+1)$:

$$\Lambda P_n^m \begin{Bmatrix} c_m \\ s_m \end{Bmatrix} = -n(n+1) P_n^m \begin{Bmatrix} c_m \\ s_m \end{Bmatrix} \quad (11)$$

The Ω - operator acting on a spherical harmonic gives a linear combination of spherical harmonics:

$$\Omega P_n^m \begin{Bmatrix} c_m \\ s_m \end{Bmatrix} = \mp \sum_{j=m-2}^{m+2} \sum_{k=n-2}^{n+2} a_{nk}^{mj} P_k^j \begin{Bmatrix} c_j \\ s_j \end{Bmatrix} \quad (12)$$

For c_m , use the minus sign, and require $m \geq 0$; for s_m , use the plus sign and require $m > 0$. The coefficients a_{nk}^{mj} are given in Table 1 of [26] (or see TABLE V in [30]); in using this table keep in mind that the table is valid only for $m \geq 0$ and that $P_n^m = 0$ if $m > n$, and that:

$$\Omega(1) = \Omega(P_0^0 c_0) = -\frac{1}{2} P_2^0 s_2 \quad (13)$$

which we will use below.

IV. BIRD-ARMSTRONG PAREN FUNCTIONS

The orientation distribution function will be given in terms of the first four of the Bird-Armstrong “paren” functions (see Eq. (10) of [16] or Eqs. (28)-(30) of [30]). A sample paren function is defined as follows:

$$(a, b, c; t) = \frac{1}{\lambda^3} \int_{-\infty}^t \int_{-\infty}^{t'} \int_{-\infty}^{t''} e^{-a(t-t')/\lambda} e^{-b(t-t'')/\lambda} e^{-c(t-t''')/\lambda} f(t') f(t'') f(t''') dt''' dt'' dt' \quad (14)$$

where a , b and c are dimensionless. The paren function above may be differentiated with respect to time by using the Leibniz formula for differentiating integrals to get:

$$\lambda \frac{d}{dt} (a, b, c; t) = -(a + b + c) (a, b, c; t) + f(t) (b, c; t) \quad (15)$$

We need these for developing the expression for the orientation distribution function.

V. EXPANSION OF THE ORIENTATION DISTRIBUTION FUNCTION FOR LAOS

The equation for the orientation distribution function given in Eq. (6), for any flow given by Eq. (1), may be solved approximately by postulating a solution of the form:

$$\psi(\theta, \phi, t) = \frac{1}{4\pi} \left[1 + (6\lambda\dot{\gamma}^0) \psi_1(\theta, \phi, t) + (6\lambda\dot{\gamma}^0)^2 \psi_2(\theta, \phi, t) + (6\lambda\dot{\gamma}^0)^3 \psi_3(\theta, \phi, t) + (6\lambda\dot{\gamma}^0)^4 \psi_4(\theta, \phi, t) + \dots \right] \quad (16)$$

where the factor $1/4\pi$ is the same normalization constant [see just before Eq. (7) above]. Thus for a fluid at rest:

$$\psi(\theta, \phi) = \frac{1}{4\pi} \quad (17)$$

When Eq. (16) is substituted into the partial differential equation for ψ , Eq. (6), and, equating equal powers of the shear rate amplitude, we get the following series of differential equations:

$$6\lambda \frac{\partial}{\partial t} \psi_1 = \Lambda \psi_1 - \Omega(1) f(t) \quad (18)$$

$$6\lambda \frac{\partial}{\partial t} \psi_2 = \Lambda \psi_2 - \Omega(\psi_1) f(t) \quad (19)$$

$$6\lambda \frac{\partial}{\partial t} \psi_3 = \Lambda \psi_3 - \Omega(\psi_2) f(t) \quad (20)$$

$$6\lambda \frac{\partial}{\partial t} \psi_4 = \Lambda \psi_4 - \Omega(\psi_3) f(t) \quad (21)$$

and so on, where for large-amplitude oscillatory shear flow:

$$\psi_1(\theta, \phi, t) = \frac{1}{12} P_2^2 s_2(1; t) \quad (22)$$

$$\psi_2(\theta, \phi, t) = -\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) (0, 1; t) - \left(\frac{6}{7} P_4^0 c_0 - \frac{1}{28} P_4^4 c_4 \right) \left(\frac{7}{3}, 1; t \right) \right] \quad (23)$$

and

$$\begin{aligned} \psi_3(\theta, \phi, t) = & -\frac{1}{10584} \left[23(0, 0, 1; t) + 12\left(-\frac{7}{3}, \frac{7}{3}, 1; t\right) \right] P_2^2 s_2 \\ & - \frac{1}{432} \left[\frac{3}{49} \left(\frac{7}{3}, 0, 1; t\right) + \frac{30}{539} \left(0, \frac{7}{3}, 1; t\right) \right] P_4^2 s_2 \\ & + \frac{1}{432(14)} \left[\frac{1}{2} \left(\frac{7}{3}, 0, 1; t\right) + \left(0, \frac{7}{3}, 1; t\right) \right] P_4^4 s_4 \\ & + \frac{1}{432(22)} \left[P_6^2 s_2 - \frac{1}{72} P_6^6 s_6 \right] \left(\frac{11}{3}, \frac{7}{3}, 1; t\right) \end{aligned} \quad (24)$$

were given in Eqs. (34), (45) and (54) of [12]. Whereas the **reddened** terms in Eq. (24) do not contribute to either the shear stress or the normal stress difference responses in large-amplitude oscillatory shear flow, they are important contributions to the shape of the orientation distribution function (see Addendum to Ref. [16]). We must thus include them when rendering the orientation distribution function in three dimensions. We next solve Eq. (21) for $\psi_4(\theta, \phi, t)$.

Solution for $\psi_4(\theta, \phi, t)$

Use of Eq. (6) and Table 1 in [26] leads to the differential equation for $\psi_4(\theta, \phi, t)$:

$$6\lambda \frac{\partial}{\partial t} \psi = \Lambda \psi_4 - f(t) \Omega \left[-\frac{1}{10584} [23(0,0,1;t) + 12(-\frac{z}{3}, \frac{z}{3}, 1;t)] P_2^2 s_2 \right. \\ \left. -\frac{1}{432} \left[\frac{3}{49} (\frac{z}{3}, 0, 1;t) + \frac{30}{539} (0, \frac{z}{3}, 1;t) \right] P_4^2 s_2 \right. \\ \left. +\frac{1}{6048} \left[\frac{1}{2} (\frac{z}{3}, 0, 1;t) + (0, \frac{z}{3}, 1;t) \right] P_4^4 s_4 \right] \quad (25)$$

The omega operator acts on the spherical harmonics appearing here to give:

$$\Omega P_2^2 s_2 = \frac{6}{7} P_2^0 c_0 - P_2^2 c_2 - \frac{6}{7} P_4^0 c_0 + \frac{1}{28} P_4^4 c_4 \quad (26)$$

$$\Omega P_4^2 s_2 = \frac{20}{7} P_2^0 c_0 + \frac{270}{77} P_4^0 c_0 - \frac{70}{11} P_6^0 c_0 - P_4^2 c_2 - \frac{3}{308} P_4^4 c_4 + P_6^4 c_4 \quad (27)$$

$$\Omega P_4^4 s_4 = +\frac{40}{3} P_2^2 c_2 + \frac{12}{11} P_4^2 c_2 - \frac{14}{33} P_6^2 c_2 - 2P_4^4 c_4 + \frac{7}{396} P_6^6 c_6 \quad (28)$$

$$6\lambda \frac{\partial}{\partial t} \psi_4 = \Lambda \psi_4 - f(t) \left[-\frac{1}{10584} [23(0,0,1;t) + 12(-\frac{z}{3}, \frac{z}{3}, 1;t)] \left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 + \dots \right) \right. \\ \left. -\frac{1}{432} \frac{3}{49} \left[(\frac{z}{3}, 0, 1;t) + \frac{10}{11} (0, \frac{z}{3}, 1;t) \right] \left(\frac{20}{7} P_2^0 c_0 + \dots \right) \right. \\ \left. +\frac{1}{6048} \left[\frac{1}{2} (\frac{z}{3}, 0, 1;t) + (0, \frac{z}{3}, 1;t) \right] \left(\frac{40}{3} P_2^2 c_2 + \dots \right) \right] \quad (29)$$

We now postulate a solution of the form:

$$\psi_4 = K_1(t) P_2^0 c_0 + K_2(t) P_2^2 c_2 + L_1(t) P_2^0 c_0 + L_2(t) P_2^2 c_2 + \dots \quad (30)$$

Then these first-order differential equations may be solved to give:

$$K_1 = \frac{1}{7\lambda} \cdot \frac{1}{10584} \int_{-\infty}^t e^{-(t-t')/\lambda} f(t') \{ [23(0,0,1;t')] + 12(-\frac{z}{3}, \frac{z}{3}, 1;t') \} dt' \\ = \frac{1}{7\lambda^4} \cdot \frac{1}{10584} \int_{-\infty}^t e^{-(t-t')/\lambda} f(t') \left\{ \begin{aligned} &23 \int_{-\infty}^{t'} \int_{-\infty}^{t''} \int_{-\infty}^{t'''} e^{-(t'-t''')/\lambda} f(t'') f(t''') f(t''''') dt'''' dt''' dt'' dt' \\ &12 \int_{-\infty}^{t'} \int_{-\infty}^{t''} \int_{-\infty}^{t'''} e^{-(7/3)(t'-t'')/\lambda} e^{-(7/3)(t'-t''')/\lambda} e^{-(t'-t''')/\lambda} \\ &\cdot f(t'') f(t''') f(t''''') dt'''' dt''' dt'' dt' \end{aligned} \right\} \quad (31)$$

$$= \frac{1}{74088} [23(0,0,0,1;t) + 12(-\frac{z}{3}, \frac{z}{3}, 0, 1;t)]$$

$$K_2 = \frac{1}{63504} [23(0,0,0,1;t) + 12(-\frac{z}{3}, \frac{z}{3}, 0, 1;t)] \quad (32)$$

$$\begin{aligned}
L_1 = & -\frac{1}{6\lambda} \frac{1}{432} \frac{3}{49} \frac{20}{7} \int_{-\infty}^t e^{-(t-t')/\lambda} f(t') \left[\left(\frac{z}{3}, 0, 1; t' \right) \right] dt' \\
& - \frac{1}{6\lambda} \frac{1}{432} \frac{3}{49} \frac{20}{7} \int_{-\infty}^t e^{-(10/3)(t-t')/\lambda} f(t') \left[\frac{10}{11} \left(0, \frac{z}{3}, 1; t' \right) \right] dt' \\
= & -\frac{1}{6\lambda^4} \frac{1}{432} \frac{3}{49} \frac{20}{7} \int_{-\infty}^t e^{-(t-t')/\lambda} f(t') \int_{-\infty}^{t'} \int_{-\infty}^{t''} \int_{-\infty}^{t'''} e^{-(7/3)(t'-t'')/\lambda} e^{-(t'-t''')/\lambda} \\
& \cdot f(t'') f(t''') f(t''''') dt'''' dt''' dt'' dt' \\
& - \frac{1}{6\lambda^4} \frac{1}{432} \frac{30}{539} \frac{20}{7} \int_{-\infty}^t e^{-(t-t')/\lambda} f(t') \int_{-\infty}^{t'} \int_{-\infty}^{t''} \int_{-\infty}^{t'''} e^{-(7/3)(t'-t'')/\lambda} e^{-(t'-t''')/\lambda} \\
& \cdot f(t'') f(t''') f(t''''') dt'''' dt''' dt'' dt' \\
= & -\frac{1}{6} \cdot \frac{1}{432} \frac{3}{49} \frac{20}{7} \left(-\frac{z}{3}, \frac{z}{3}, 0, 1; t \right) - \frac{1}{6} \cdot \frac{1}{432} \frac{3}{49} \frac{10}{11} \frac{20}{7} \left(-\frac{z}{3}, 0, \frac{z}{3}, 1; t \right) \\
L_2 = & -\frac{1}{6\lambda} \frac{1}{6048} \frac{40}{3} \int_{-\infty}^t e^{-(t-t')/\lambda} f(t') \left[\frac{1}{2} \left(\frac{z}{3}, 0, 1; t \right) \right] dt' \\
& - \frac{1}{6\lambda} \frac{1}{3 \cdot 6048} \frac{40}{3} \int_{-\infty}^t e^{-(t-t')/\lambda} f(t') \left[\left(0, \frac{z}{3}, 1; t \right) \right] dt'
\end{aligned} \tag{33}$$

$$\begin{aligned}
= & -\frac{1}{6} \cdot \frac{1}{6048} \cdot \frac{1}{2} \frac{40}{3} \left(-\frac{z}{3}, \frac{z}{3}, 0, 1; t \right) - \frac{1}{6} \cdot \frac{1}{6048} \frac{40}{3} \left(-\frac{z}{3}, 0, \frac{z}{3}, 1; t \right) \\
L_2 = & -\frac{1}{6\lambda} \frac{1}{6048} \frac{40}{3} \int_{-\infty}^t e^{-(t-t')/\lambda} f(t') \left[\frac{1}{2} \left(\frac{z}{3}, 0, 1; t \right) \right] dt' \\
& - \frac{1}{6\lambda} \frac{1}{3 \cdot 6048} \frac{40}{3} \int_{-\infty}^t e^{-(t-t')/\lambda} f(t') \left[\left(0, \frac{z}{3}, 1; t \right) \right] dt' \\
= & -\frac{1}{6} \cdot \frac{1}{6048} \cdot \frac{1}{2} \frac{40}{3} \left(-\frac{z}{3}, \frac{z}{3}, 0, 1; t \right) - \frac{1}{6} \cdot \frac{1}{6048} \frac{40}{3} \left(-\frac{z}{3}, 0, \frac{z}{3}, 1; t \right)
\end{aligned} \tag{34}$$

and therefore, in large-amplitude oscillatory shear flow, ψ_4 is given by:

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

Knowing Eqs. (22), (23), (24), and (35), we can solve for the orientation distribution function up to including terms up to ψ_4 .

a. FOURIER SERIES FOR ψ_1

By substituting Eq. (71) from Section IX into Eq. (22), and the associated Legendre polynomial from Table III into Eq. (22), we get:

$$\psi_1(\theta, \phi, t) = \frac{1}{4} (1 - C^2) s_2 \left(\frac{\cos \omega t + \lambda \omega \sin \omega t}{1 + \Lambda} \right) \tag{36}$$

which, in the limit as $\lambda \omega \rightarrow 0$, reduces to:

$$\psi_1(\theta, \phi, t) = \frac{1}{4} (1 - C^2) s_2 \tag{37}$$

which applies to steady shear flow.

b. FOURIER SERIES FOR ψ_2

By substituting Eqs. (72) and (73) from Section IX into Eq. (23), and the associated Legendre polynomial from Table III into the result, we get:

$$\psi_2(\theta, \phi, t) = -\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) \left[\frac{(1-2\Lambda) \cos 2\omega t + 3\lambda \omega \sin 2\omega t}{2(1+4\Lambda)(1+\Lambda)} + \frac{1}{2(1+\Lambda)} \right] - \left(\frac{6}{7} P_4^0 c_0 - \frac{1}{28} P_4^4 c_4 \right) \left[\frac{3(10-6\Lambda) \cos 2\omega t + 48\lambda \omega \sin 2\omega t}{16(25+9\Lambda)(1+\Lambda)} + \frac{3}{20(1+\Lambda)} \right] \right] \quad (38)$$

which can be separated into the different harmonics of ωt :

$$\psi_2(\theta, \phi, t) = -\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 \right) \frac{(1-2\Lambda) \cos 2\omega t + 3\lambda \omega \sin 2\omega t}{2(1+4\Lambda)(1+\Lambda)} + \left(\frac{6}{7} P_2^0 c_0 \right) \frac{1}{2(1+\Lambda)} + \left(-P_2^2 c_2 \right) \frac{(1-2\Lambda) \cos 2\omega t + 3\lambda \omega \sin 2\omega t}{2(1+4\Lambda)(1+\Lambda)} + \left(-P_2^2 c_2 \right) \frac{1}{2(1+\Lambda)} + \left(-\frac{6}{7} P_4^0 c_0 \right) \frac{3(10-6\Lambda) \cos 2\omega t + 48\lambda \omega \sin 2\omega t}{16(25+9\Lambda)(1+\Lambda)} + \left(-\frac{6}{7} P_4^0 c_0 \right) \frac{3}{20(1+\Lambda)} + \left(\frac{1}{28} P_4^4 c_4 \right) \frac{3(10-6\Lambda) \cos 2\omega t + 48\lambda \omega \sin 2\omega t}{16(25+9\Lambda)(1+\Lambda)} + \left(\frac{1}{28} P_4^4 c_4 \right) \frac{3}{20(1+\Lambda)} \right] \quad (39)$$

where we separate it into the different harmonics for ease of manipulation. Each harmonic of ψ_2 is denoted by a subscript, $ck \equiv \cos k\omega t$ or $sk \equiv \sin k\omega t$:

$$\psi_2(\theta, \phi, t)_{c2} = -\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) \frac{(1-2\Lambda)}{2(1+4\Lambda)(1+\Lambda)} + \left(\frac{1}{28} P_4^4 c_4 - \frac{6}{7} P_4^0 c_0 \right) \frac{3(10-6\Lambda)}{16(25+9\Lambda)(1+\Lambda)} \right] \cos 2\omega t \quad (40)$$

$$\psi_2(\theta, \phi, t)_{s2} = -\frac{1}{72} \left[\left(\frac{6}{7} P_2^0 c_0 - P_2^2 c_2 \right) \frac{3}{2(1+4\Lambda)(1+\Lambda)} + \left(\frac{1}{28} P_4^4 c_4 - \frac{6}{7} P_4^0 c_0 \right) \frac{48}{16(25+9\Lambda)(1+\Lambda)} \right] \lambda \omega \sin 2\omega t \quad (41)$$

$$\psi_2(\theta, \phi, t)_{c0} = -\frac{1}{72} \left[\frac{\frac{6}{7} P_2^0 c_0 - P_2^2 c_2}{2(1+\Lambda)} + \frac{\frac{3}{28} P_4^4 c_4 - \frac{18}{7} P_4^0 c_0}{20(1+\Lambda)} \right] \quad (42)$$

and by summing Eq. (40) through Eq. (42), ψ_2 becomes:

$$\psi_2(\theta, \phi, t) = -\frac{1}{72} \left\{ \left[\frac{\left(\frac{48}{7} P_2^0 c_0 - 8P_2^2 c_2 \right) (1-2\Lambda)(25+9\Lambda) + \left(\frac{3}{28} P_4^4 c_4 - \frac{18}{7} P_4^0 c_0 \right) (10-6\Lambda)(1+4\Lambda)}{16(1+\Lambda)(1+4\Lambda)(25+9\Lambda)} \right] \cos 2\omega t + \left[\frac{\left(\frac{18}{7} P_2^0 c_0 - 3P_2^2 c_2 \right) (25+9\Lambda) + \left(\frac{6}{28} P_4^4 c_4 - \frac{36}{7} P_4^0 c_0 \right) (1+4\Lambda)}{2(1+\Lambda)(1+4\Lambda)(25+9\Lambda)} \right] \lambda \omega \sin 2\omega t + \frac{\frac{60}{7} P_2^0 c_0 - 10P_2^2 c_2 + \frac{3}{28} P_4^4 c_4 - \frac{18}{7} P_4^0 c_0}{20(1+\Lambda)} \right\} \quad (43)$$

We now substitute the relevant associated Legendre polynomials from Table III, which gives:

$$\psi_2(\theta, \phi, t) = -\frac{1}{72} \left\{ \begin{aligned} & \left[\begin{aligned} & \frac{24}{7}(3C^2-1)(1-2\Lambda)(25+9\Lambda) \\ & -\frac{9}{28}(35C^4-30C^2+3)(10-6\Lambda)(1+4\Lambda) \\ & -24(1-C^2)(1-2\Lambda)(25+9\Lambda)c_2 \\ & +\frac{315}{28}(1-C^2)^2(10-6\Lambda)(1+4\Lambda)c_4 \end{aligned} \right] \cos 2\omega t \\ & + \left[\begin{aligned} & \frac{9}{7}(3C^2-1)(25+9\Lambda) - \frac{9}{14}(35C^4-30C^2+3)(1+4\Lambda) \\ & -9(1-C^2)(25+9\Lambda)c_2 + \frac{630}{28}(1-C^2)^2(1+4\Lambda)c_4 \end{aligned} \right] \lambda\omega \sin 2\omega t \\ & + \left[\begin{aligned} & -\frac{3}{4}(15C^4+30C^2-7) - 30(1-C^2)c_2 + \frac{315}{28}(1-C^2)^2c_4 \end{aligned} \right] \end{aligned} \right\} \quad (44)$$

which, in the limit as $\lambda\omega \rightarrow 0$, reduces to:

$$\psi_2(\theta, \phi, t) = -\frac{1}{144} \left[-\frac{135}{8}C^4 - \frac{123}{28}C^2 + \frac{19}{56} - 61(1-C^2)c_2 + \frac{135}{8}(1-C^2)^2c_4 \right] \quad (45)$$

which applies to steady shear flow.

c. FOURIER SERIES FOR ψ_3

Eq. (24) here can be rewritten as:

$$\begin{aligned} \psi_3(\theta, \phi, t) = & -\frac{23}{10584}P_2^2s_2(0,0,1;t) \\ & -\frac{1}{882}P_2^2s_2\left(-\frac{z}{3}, \frac{z}{3}, 1; t\right) \\ & + \left[\frac{1}{12096}P_4^4s_4 - \frac{1}{7056}P_4^2s_2 \right] \left(\frac{z}{3}, 0, 1; t\right) \\ & + \left[\frac{1}{432(14)}P_4^4s_4 - \frac{1}{432} \frac{30}{539}P_4^2s_2 \right] \left(0, \frac{z}{3}, 1; t\right) \\ & + \frac{1}{432(22)} \left[P_6^2s_2 - \frac{1}{72}P_6^6s_6 \right] \left(\frac{11}{3}, \frac{z}{3}, 1; t\right) \end{aligned} \quad (46)$$

and by substituting Eqs. (76), (77), (78), (79), and (80) from Section X into (46), we get:

$$\begin{aligned}
\psi_3(\theta, \phi, t) = & -\frac{23}{10584} P_2^2 S_2 \left[\frac{(1-11\Lambda) \cos 3\omega t + 6(1-\Lambda)\lambda\omega \sin 3\omega t}{4(1+9\Lambda)(1+4\Lambda)(1+\Lambda)} \right. \\
& \left. + \frac{3 \cos \omega t + 6\lambda\omega \sin \omega t}{4(1+4\Lambda)(1+\Lambda)} \right] \\
& - \frac{1}{882} P_2^2 S_2 \left[\frac{3(5-27\Lambda) \cos 3\omega t + 3(23-9\Lambda)\lambda\omega \sin 3\omega t}{8(1+9\Lambda)(25+9\Lambda)(1+\Lambda)} \right. \\
& \left. + \frac{3(75-37\Lambda) \cos \omega t + 3(3\Lambda+115)\lambda\omega \sin \omega t}{40(25+9\Lambda)(1+\Lambda)^2} \right] \\
& + \left[\frac{1}{12096} P_4^4 S_4 - \frac{1}{7056} P_4^2 S_2 \right] \left[\frac{3(10-47\Lambda) \cos 3\omega t + 9(13-6\Lambda)\lambda\omega \sin 3\omega t}{4(1+4\Lambda)(100+81\Lambda)(1+\Lambda)} \right. \\
& \left. + \frac{9(10+17\Lambda) \cos \omega t + 9(13+6\Lambda)\lambda\omega \sin \omega t}{4(1+4\Lambda)(100+9\Lambda)(1+\Lambda)} \right] \\
& + \left[\frac{1}{432(14)} P_4^4 S_4 - \frac{1}{432} \frac{30}{539} P_4^2 S_2 \right] \left[\frac{18(25-51\Lambda) \cos 3\omega t + 9(125-27\Lambda)\lambda\omega \sin 3\omega t}{8(25+9\Lambda)(100+81\Lambda)(1+\Lambda)} \right. \\
& \left. + \frac{270(25-3\Lambda) \cos \omega t + 9(625+9\Lambda)\lambda\omega \sin \omega t}{40(1+4\Lambda)(100+9\Lambda)(1+\Lambda)} \right] \\
& + \frac{1}{432(22)} \left[P_6^2 S_2 - \frac{1}{72} P_6^6 S_6 \right] \left[\frac{15(7-9\Lambda) \cos 3\omega t + 3(71-9\Lambda)\lambda\omega \sin 3\omega t}{8(25+9\Lambda)(49+9\Lambda)(1+\Lambda)} \right. \\
& \left. + \frac{3(525-19\Lambda) \cos \omega t + 3(355+3\Lambda)\lambda\omega \sin \omega t}{40(25+9\Lambda)(1+\Lambda)(49+\Lambda)} \right]
\end{aligned} \tag{47}$$

which we can break up into the different harmonics as in Section V.b:

$$\psi_3(\theta, \phi, t)_{c3} = \left\{ \begin{aligned} & \left[\frac{603\Lambda^2 + 892\Lambda - 95}{6048(\Lambda+1)(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)} \right] P_2^2 S_2 \\ & + \left[\frac{19\Lambda - 5}{14784(\Lambda+1)(4\Lambda+1)(9\Lambda+25)} \right] P_4^2 S_2 \\ & + \left[\frac{5(7-9\Lambda)}{25344(\Lambda+1)(9\Lambda+25)(9\Lambda+49)} \right] P_6^2 S_2 \\ & - \left[\frac{1647\Lambda^2 + 791\Lambda - 400}{16128(\Lambda+1)(4\Lambda+1)(9\Lambda+25)(81\Lambda+100)} \right] P_4^4 S_4 \\ & + \left[\frac{5(9\Lambda-7)}{1824768(\Lambda+1)(9\Lambda+25)(9\Lambda+49)} \right] P_6^6 S_6 \end{aligned} \right\} \cos 3\omega t \tag{48}$$

$$\psi_3(\theta, \phi, t)_{s_3} = \left\{ \begin{aligned} & \left[\frac{45\Lambda^2 + 17\Lambda - 92}{1008(\Lambda+1)(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)} \right] P_2^2 s_2 \\ & + \left[\frac{\Lambda - 3}{2464(\Lambda+1)(4\Lambda+1)(9\Lambda+25)} \right] P_4^2 s_2 \\ & + \left[\frac{71 - 9\Lambda}{25344(9\Lambda+25)(9\Lambda+49)(\Lambda+1)} \right] P_6^2 s_2 \\ & - \left[\frac{81\Lambda^2 - 220\Lambda - 225}{2688(\Lambda+1)(4\Lambda+1)(9\Lambda+25)(81\Lambda+100)} \right] P_4^4 s_4 \\ & + \left[\frac{9\Lambda - 71}{1824768(\Lambda+1)(9\Lambda+25)(9\Lambda+49)} \right] P_6^6 s_6 \end{aligned} \right\} \lambda \omega \sin 3\omega t \quad (49)$$

$$\psi_3(\theta, \phi, t)_{c1} = \left\{ \begin{aligned} & \left[-\frac{75 - 37\Lambda}{11760(\Lambda+1)^2(9\Lambda+25)} \right] P_2^2 s_2 \\ & - \left[\frac{97\Lambda + 860}{34496(\Lambda+1)(4\Lambda+1)(9\Lambda+100)} \right] P_4^2 s_2 \\ & + \left[\frac{525 - 19\Lambda}{126720(\Lambda+1)(\Lambda+49)(9\Lambda+25)} \right] P_6^2 s_2 \\ & + \left[\frac{160 - \Lambda}{5376(\Lambda+1)(4\Lambda+1)(9\Lambda+100)} \right] P_4^4 s_4 \\ & - \left[\frac{525 - 19\Lambda}{9123840(\Lambda+1)(\Lambda+49)(9\Lambda+25)} \right] P_6^6 s_6 \end{aligned} \right\} \cos \omega t \quad (50)$$

$$\psi_3(\theta, \phi, t)_{s1} = \left\{ \begin{aligned} & - \left[\frac{153\Lambda^2 + 757\Lambda + 460}{5040(\Lambda+1)^2(4\Lambda+1)(9\Lambda+25)} \right] P_2^2 s_2 \\ & - \left[\frac{3(25\Lambda + 256)}{34496(\Lambda+1)(4\Lambda+1)(9\Lambda+100)} \right] P_4^2 s_2 \\ & + \left[\frac{3\Lambda + 355}{126720(9\Lambda+25)(\Lambda+1)(\Lambda+49)} \right] P_6^2 s_2 \\ & + \left[\frac{13\Lambda + 230}{8960(\Lambda+1)(4\Lambda+1)(9\Lambda+100)} \right] P_4^4 s_4 \\ & - \left[\frac{3\Lambda + 355}{9123840(\Lambda+1)(\Lambda+49)(9\Lambda+25)} \right] P_6^6 s_6 \end{aligned} \right\} \lambda \omega \sin \omega t \quad (51)$$

and by substituting in the associated Legendre polynomials from Table III leads to:

$$\begin{aligned}
\psi_3(\theta, \phi, t) = & \left[\begin{aligned} & \left[\begin{aligned} & \frac{1}{2016}(603\Lambda^2 + 892\Lambda - 95)(9\Lambda + 49)(81\Lambda + 100) \\ & + \frac{5}{9856}(19\Lambda - 5)(9\Lambda + 1)(9\Lambda + 49)(81\Lambda + 100)(7C^2 - 1) \\ & + \frac{36}{13903}(7 - 9\Lambda)(4\Lambda + 1)(9\Lambda + 1)(81\Lambda + 100)(33C^4 - 18C^2 + 1) \end{aligned} \right] s_2 \\ & - \frac{5}{768}(1647\Lambda^2 + 791\Lambda - 400)(9\Lambda + 1)(9\Lambda + 49)(1 - C^2)s_4 \\ & + \frac{175}{6144}(9\Lambda - 7)(4\Lambda + 1)(9\Lambda + 1)(81\Lambda + 100)(1 - C^2)^2 s_6 \end{aligned} \right] \frac{(1 - C^2)}{(\Lambda + 1)(4\Lambda + 1)(9\Lambda + 1)(9\Lambda + 25)(9\Lambda + 49)(81\Lambda + 100)} \cos 3\omega t \\ & + \left[\begin{aligned} & \left[\begin{aligned} & \frac{1}{336}(9\Lambda + 49)(81\Lambda + 100)(45\Lambda^2 + 17\Lambda - 92) \\ & + \frac{15}{4928}(\Lambda - 3)(9\Lambda + 1)(9\Lambda + 49)(81\Lambda + 100)(7C^2 - 1) \\ & + \frac{35}{67584}(71 - 9\Lambda)(4\Lambda + 1)(9\Lambda + 1)(81\Lambda + 100)(33C^4 - 18C^2 + 1) \end{aligned} \right] s_2 \\ & - \frac{5}{128}(81\Lambda^2 - 220\Lambda - 225)(9\Lambda + 1)(9\Lambda + 49)(1 - C^2)s_4 \\ & + \frac{35}{6144}(9\Lambda - 71)(4\Lambda + 1)(9\Lambda + 1)(81\Lambda + 100)(1 - C^2)^2 s_6 \end{aligned} \right] \frac{(1 - C^2)}{(\Lambda + 1)(4\Lambda + 1)(9\Lambda + 1)(9\Lambda + 25)(9\Lambda + 49)(81\Lambda + 100)} \lambda\omega \sin 3\omega t \\ & + \left[\begin{aligned} & \left[\begin{aligned} & -\frac{1}{3920}(75 - 37\Lambda)(\Lambda + 49)(4\Lambda + 1)(9\Lambda + 100) \\ & - \frac{15}{68992}(97\Lambda + 860)(\Lambda + 1)(\Lambda + 49)(9\Lambda + 25)(7C^2 - 1) \\ & + \frac{7}{67584}(525 - 19\Lambda)(\Lambda + 1)(4\Lambda + 1)(9\Lambda + 100)(33C^4 - 18C^2 + 1) \end{aligned} \right] s_2 \\ & + \frac{5}{256}(160 - \Lambda)(\Lambda + 1)(\Lambda + 49)(9\Lambda + 25)(1 - C^2)s_4 \\ & - \frac{7}{6144}(525 - 19\Lambda)(\Lambda + 1)(4\Lambda + 1)(9\Lambda + 100)(1 - C^2)^2 s_6 \end{aligned} \right] \frac{(1 - C^2)}{(\Lambda + 1)^2(\Lambda + 49)(4\Lambda + 1)(9\Lambda + 25)(9\Lambda + 100)} \cos \omega t \\ & + \left[\begin{aligned} & \left[\begin{aligned} & -\frac{1}{1680}(\Lambda + 49)(9\Lambda + 100)(153\Lambda^2 + 757\Lambda + 460) \\ & - \frac{45}{68992}(25\Lambda + 256)(\Lambda + 1)(\Lambda + 49)(9\Lambda + 25)(7C^2 - 1) \\ & + \frac{7}{67584}(3\Lambda + 355)(\Lambda + 1)(4\Lambda + 1)(9\Lambda + 100)(33C^4 - 18C^2 + 1) \end{aligned} \right] s_2 \\ & + \frac{3}{256}(13\Lambda + 230)(\Lambda + 1)(\Lambda + 49)(9\Lambda + 25)(1 - C^2)s_4 \\ & - \frac{7}{6144}(3\Lambda + 355)(\Lambda + 1)(4\Lambda + 1)(9\Lambda + 100)(1 - C^2)^2 s_6 \end{aligned} \right] \frac{(1 - C^2)}{(\Lambda + 1)^2(\Lambda + 49)(4\Lambda + 1)(9\Lambda + 25)(9\Lambda + 100)} \lambda\omega \sin \omega t \end{aligned} \right]
\end{aligned} \tag{52}$$

which is the expression for ψ_3 in large-amplitude oscillatory shear flow. In the limit as $\lambda\omega \rightarrow 0$, Eq. (52) reduces to:

$$\psi_3(\theta, \phi, t) = \frac{1}{122500} \left[\begin{aligned} & \left[\begin{aligned} & \frac{1703117325}{7118336}C^4 - \frac{14076786975}{39150848}C^2 + \frac{254417947975}{704715264} \end{aligned} \right] s_2 \\ & + \frac{189875}{48}(1 - C^2)s_4 - \frac{30625}{384}(1 - C^2)^2 s_6 \end{aligned} \right] (1 - C^2) \tag{53}$$

which applies to steady shear flow.

d. FOURIER SERIES FOR ψ_4

Eq. (35) can be rewritten as:

$$\begin{aligned}\psi_4(\theta, \phi, t) = & \left[-\frac{23}{74088} P_2^0 c_0 - \frac{23}{63504} P_2^2 c_2 \right] (0, 0, 0, 1; t) \\ & + \left[-\frac{1}{6174} P_2^0 c_0 - \frac{1}{5292} P_2^2 c_2 \right] \left(0, -\frac{7}{3}, \frac{7}{3}, 1; t \right) \\ & + \left[-\frac{5}{74088} P_2^0 c_0 - \frac{5}{27216} P_2^2 c_2 \right] \left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t \right) \\ & + \left[-\frac{25}{407484} P_2^0 c_0 - \frac{5}{13608} P_2^2 c_2 \right] \left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t \right)\end{aligned}\quad (54)$$

and by substituting Eqs. (83), (84), (85), and (86) from Section XI into (46), we get:

$$\begin{aligned}\psi_4(\theta, \phi, t) = & \left[-\frac{23}{74088} P_2^0 c_0 - \frac{23}{63504} P_2^2 c_2 \right] \left[\frac{1}{8} \frac{(1-35\Lambda+24\Lambda^2)\cos 4\omega t + (10-50\Lambda)\lambda\omega \sin 4\omega t}{(1+16\Lambda)(1+9\Lambda)(1+4\Lambda)(1+\Lambda)} \right. \\ & \left. + \frac{1}{8} \frac{\left[(4-13\Lambda-32\Lambda^2-159\Lambda^3)\cos 2\omega t + 2(10+26\Lambda+61\Lambda^2-27\Lambda^3)\lambda\omega \sin 2\omega t \right]}{(1+4\Lambda)(1+9\Lambda)(1+\Lambda)^3} \right. \\ & \left. + \frac{3}{8} \frac{1}{(1+4\Lambda)(1+\Lambda)} \right] \\ & + \left[-\frac{1}{6174} P_2^0 c_0 - \frac{1}{5292} P_2^2 c_2 \right] \left[\frac{3}{4} \frac{(5-119\Lambda+36\Lambda^2)\cos 4\omega t + (43-117\Lambda)\lambda\omega \sin 4\omega t}{(1+16\Lambda)(1+9\Lambda)(100+36\Lambda)(1+\Lambda)} \right. \\ & \left. + \frac{3}{20} \frac{\left[(100+243\Lambda-2289\Lambda^2-5915\Lambda^3-1179\Lambda^4)\cos 2\omega t + 2(215+1447\Lambda+1934\Lambda^2-693\Lambda^3-243\Lambda^4)\lambda\omega \sin 2\omega t \right]}{(1+4\Lambda)(1+9\Lambda)(100+36\Lambda)(1+\Lambda)^3} \right. \\ & \left. + \frac{3}{20} \frac{(75-37\Lambda)}{(1+\Lambda)^2(100+36\Lambda)} \right] \\ & + \left[-\frac{5}{74088} P_2^0 c_0 - \frac{5}{27216} P_2^2 c_2 \right] \left[\frac{3}{8} \frac{(10-203\Lambda+72\Lambda^2)\cos 4\omega t + (79-206\Lambda)\lambda\omega \sin 4\omega t}{(1+16\Lambda)(100+81\Lambda)(1+4\Lambda)(1+\Lambda)} \right. \\ & \left. + \frac{3}{8} \frac{\left[(4000-5680\Lambda-10262\Lambda^2-38031\Lambda^3-30780\Lambda^4)\cos 2\omega t + (15800+35050\Lambda+63881\Lambda^2+32130\Lambda^3-5832\Lambda^4)\lambda\omega \sin 2\omega t \right]}{(1+16\Lambda)(100+81\Lambda)(1+4\Lambda)(1+\Lambda)^2(100+9\Lambda)} \right. \\ & \left. + \frac{9}{8} \frac{(10+17\Lambda)}{(100+9\Lambda)(1+4\Lambda)(1+\Lambda)} \right] \\ & + \left[-\frac{25}{407484} P_2^0 c_0 - \frac{5}{13608} P_2^2 c_2 \right] \left[\frac{9}{4} \frac{2(25-301\Lambda+54\Lambda^2)\cos 4\omega t + 5(65-87\Lambda)\lambda\omega \sin 4\omega t}{(1+16\Lambda)(100+81\Lambda)(100+36\Lambda)(1+\Lambda)} \right. \\ & \left. + \frac{9}{20} \frac{\left[(100000-72000\Lambda-475930\Lambda^2-330813\Lambda^3-62451\Lambda^4)\cos 2\omega t + (325000+622150\Lambda+269979\Lambda^2-4725\Lambda^3-13122\Lambda^4)\lambda\omega \sin 2\omega t \right]}{(1+4\Lambda)(100+81\Lambda)(100+36\Lambda)(1+\Lambda)^2(100+9\Lambda)} \right. \\ & \left. + \frac{27}{2} \frac{(25-3\Lambda)}{(100+9\Lambda)(100+36\Lambda)(1+\Lambda)} \right]\end{aligned}\quad (55)$$

which we can break up into the different harmonics as in Section V.b as:

$$\psi_4(\theta, \phi, t)_{c4} = \left\{ \begin{aligned} & - \left[\frac{5(216\Lambda^3 - 201\Lambda^2 - 497\Lambda + 16)}{66528(\Lambda+1)(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(16\Lambda+1)} \right] P_2^0 c_0 \\ & - \left[\frac{5(17496\Lambda^4 - 20979\Lambda^3 - 58383\Lambda^2 - 31526\Lambda + 1150)}{36288(\Lambda+1)(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(16\Lambda+1)(81\Lambda+100)} \right] P_2^2 c_2 \end{aligned} \right\} \cos 4\omega t \quad (56)$$

$$\psi_4(\theta, \phi, t)_{s4} = \left\{ \begin{aligned} & \left[\frac{18549162\Lambda^3 + 21819195\Lambda^2 + 7990963\Lambda - 3933950}{3259872(\Lambda+1)(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(16\Lambda+1)(81\Lambda+100)} \right] P_2^0 c_0 \\ & + \left[\frac{737586\Lambda^3 + 160560\Lambda^2 - 29711\Lambda - 63725}{36288(\Lambda+1)(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(16\Lambda+1)(81\Lambda+100)} \right] P_2^2 c_2 \end{aligned} \right\} \lambda\omega \sin 4\omega t \quad (57)$$

$$\psi_4(\theta, \phi, t)_{c2} = \left\{ \begin{aligned} & \left[\frac{\begin{pmatrix} 27917501148\Lambda^7 + 395970769863\Lambda^6 + 1359686945763\Lambda^5 \\ + 1401100327677\Lambda^4 + 389057165809\Lambda^3 + 45722628300\Lambda^2 \\ - 22856340000\Lambda - 1568000000 \end{pmatrix}}{32598720(\Lambda+1)^3(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(9\Lambda+100)(16\Lambda+1)(81\Lambda+100)} \right] P_2^0 c_0 \\ & + \left[\frac{\begin{pmatrix} 506797884\Lambda^7 + 6027824709\Lambda^6 + 19477775889\Lambda^5 \\ + 19601882811\Lambda^4 + 5251665787\Lambda^3 + 424722600\Lambda^2 \\ - 340140000\Lambda - 23000000 \end{pmatrix}}{362880(\Lambda+1)^3(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(16\Lambda+1)(81\Lambda+100)} \right] P_2^2 c_2 \end{aligned} \right\} \cos 2\omega t \quad (58)$$

$$\psi_4(\theta, \phi, t)_{s2} = \left\{ \begin{aligned} & \left[\frac{\begin{pmatrix} 4554095076\Lambda^7 + 54922330017\Lambda^6 + 62656044453\Lambda^5 \\ - 366082940052\Lambda^4 - 649050340979\Lambda^3 - 299118407355\Lambda^2 \\ - 75936742500\Lambda - 3787700000 \end{pmatrix}}{16299360(\Lambda+1)^3(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(9\Lambda+100)(16\Lambda+1)(81\Lambda+100)} \right] P_2^0 c_0 \\ & + \left[\frac{\begin{pmatrix} 73929348\Lambda^7 + 725310531\Lambda^6 + 487357884\Lambda^5 \\ - 5792477481\Lambda^4 - 9601088132\Lambda^3 - 4427460420\Lambda^2 \\ - 1076466750\Lambda - 53975000 \end{pmatrix}}{181440(\Lambda+1)^3(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(9\Lambda+100)(16\Lambda+1)(81\Lambda+100)} \right] P_2^2 c_2 \end{aligned} \right\} \lambda\omega \sin 2\omega t \quad (59)$$

$$\begin{aligned}
\psi_4(\theta, \phi, t) = & \left\{ \begin{aligned} & - \left[\frac{5(216\Lambda^3 - 201\Lambda^2 - 497\Lambda + 16)}{66528(\Lambda+1)(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(16\Lambda+1)} \right] P_2^0 c_0 \\ & - \left[\frac{5(17496\Lambda^4 - 20979\Lambda^3 - 58383\Lambda^2 - 31526\Lambda + 1150)}{36288(\Lambda+1)(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(16\Lambda+1)(81\Lambda+100)} \right] P_2^2 c_2 \end{aligned} \right\} \cos 4\omega t \\
& + \left\{ \begin{aligned} & \left[\frac{18549162\Lambda^3 + 21819195\Lambda^2 + 7990963\Lambda - 3933950}{3259872(\Lambda+1)(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(16\Lambda+1)(81\Lambda+100)} \right] P_2^0 c_0 \\ & + \left[\frac{737586\Lambda^3 + 160560\Lambda^2 - 29711\Lambda - 63725}{36288(\Lambda+1)(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(16\Lambda+1)(81\Lambda+100)} \right] P_2^2 c_2 \end{aligned} \right\} \lambda \omega \sin 4\omega t \\
& + \left\{ \begin{aligned} & \left[\frac{\begin{pmatrix} 27917501148\Lambda^7 + 395970769863\Lambda^6 + 1359686945763\Lambda^5 \\ + 1401100327677\Lambda^4 + 389057165809\Lambda^3 + 45722628300\Lambda^2 \\ - 22856340000\Lambda - 1568000000 \end{pmatrix}}{32598720(\Lambda+1)^3(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(9\Lambda+100)(16\Lambda+1)(81\Lambda+100)} \right] P_2^0 c_0 \\ & + \left[\frac{\begin{pmatrix} 506797884\Lambda^7 + 6027824709\Lambda^6 + 19477775889\Lambda^5 \\ + 19601882811\Lambda^4 + 5251665787\Lambda^3 + 424722600\Lambda^2 \\ - 340140000\Lambda - 23000000 \end{pmatrix}}{362880(\Lambda+1)^3(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(16\Lambda+1)(81\Lambda+100)} \right] P_2^2 c_2 \end{aligned} \right\} \cos 2\omega t \\
& + \left\{ \begin{aligned} & \left[\frac{\begin{pmatrix} 4554095076\Lambda^7 + 54922330017\Lambda^6 + 62656044453\Lambda^5 \\ - 366082940052\Lambda^4 - 649050340979\Lambda^3 - 299118407355\Lambda^2 \\ - 75936742500\Lambda - 3787700000 \end{pmatrix}}{16299360(\Lambda+1)^3(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(9\Lambda+100)(16\Lambda+1)(81\Lambda+100)} \right] P_2^0 c_0 \\ & + \left[\frac{\begin{pmatrix} 73929348\Lambda^7 + 725310531\Lambda^6 + 487357884\Lambda^5 \\ - 5792477481\Lambda^4 - 9601088132\Lambda^3 - 4427460420\Lambda^2 \\ - 1076466750\Lambda - 53975000 \end{pmatrix}}{181440(\Lambda+1)^3(4\Lambda+1)(9\Lambda+1)(9\Lambda+25)(9\Lambda+100)(16\Lambda+1)(81\Lambda+100)} \right] P_2^2 c_2 \end{aligned} \right\} \lambda \omega \sin 2\omega t \\
& + \left\{ \begin{aligned} & - \left[\frac{(1161\Lambda^3 + 14769\Lambda^2 + 74200\Lambda + 40000)}{110880(\Lambda+1)^2(4\Lambda+1)(9\Lambda+25)(9\Lambda+100)} \right] P_2^0 c_0 \\ & - \left[\frac{(1107\Lambda^3 + 19303\Lambda^2 + 60050\Lambda + 28750)}{60480(\Lambda+1)^2(4\Lambda+1)(9\Lambda+25)(9\Lambda+100)} \right] P_2^2 c_2 \end{aligned} \right\}
\end{aligned} \tag{60}$$

and by substituting the associated Legendre polynomials from Table III into Eqs. (56) through (60), and then summing them together, we get:

$$\begin{aligned}
\psi_4(\theta, \phi, t) = & \left\{ \frac{5(216\Lambda^3 - 201\Lambda^2 - 497\Lambda + 16)(3C^2 - 1)}{133056(\Lambda + 1)(4\Lambda + 1)(9\Lambda + 1)(9\Lambda + 25)(16\Lambda + 1)} \right\} \cos 4\omega t \\
& + \left\{ \frac{5(17496\Lambda^4 - 20979\Lambda^3 - 58383\Lambda^2 - 31526\Lambda + 1150)(1 - C^2)c_2}{12096(\Lambda + 1)(4\Lambda + 1)(9\Lambda + 1)(9\Lambda + 25)(16\Lambda + 1)(81\Lambda + 100)} \right. \\
& + \left. \frac{(18549162\Lambda^3 + 21819195\Lambda^2 + 7990963\Lambda - 3933950)(3C^2 - 1)}{6519744(\Lambda + 1)(4\Lambda + 1)(9\Lambda + 1)(9\Lambda + 25)(16\Lambda + 1)(81\Lambda + 100)} \right. \\
& + \left. \frac{(737586\Lambda^3 + 160560\Lambda^2 - 29711\Lambda - 63725)(1 - C^2)c_2}{12096(\Lambda + 1)(4\Lambda + 1)(9\Lambda + 1)(9\Lambda + 25)(16\Lambda + 1)(81\Lambda + 100)} \right\} \lambda\omega \sin 4\omega t \\
& + \left\{ \frac{\left(\begin{aligned} & 27917501148\Lambda^7 + 395970769863\Lambda^6 + 1359686945763\Lambda^5 \\ & + 1401100327677\Lambda^4 + 389057165809\Lambda^3 + 45722628300\Lambda^2 \\ & - 22856340000\Lambda - 1568000000 \end{aligned} \right) (3C^2 - 1)}{6519744(\Lambda + 1)^3(4\Lambda + 1)(9\Lambda + 1)(9\Lambda + 25)(9\Lambda + 100)(16\Lambda + 1)(81\Lambda + 100)} \right. \\
& + \left. \frac{\left(\begin{aligned} & 506797884\Lambda^7 + 6027824709\Lambda^6 + 1947775889\Lambda^5 \\ & + 19601882811\Lambda^4 + 5251665787\Lambda^3 + 424722600\Lambda^2 \\ & - 340140000\Lambda - 23000000 \end{aligned} \right) (1 - C^2)c_2}{120960(\Lambda + 1)^3(4\Lambda + 1)(9\Lambda + 1)(9\Lambda + 25)(16\Lambda + 1)(81\Lambda + 100)} \right\} \cos 2\omega t \\
& + \left\{ \frac{\left(\begin{aligned} & 4554095076\Lambda^7 + 54922330017\Lambda^6 + 62656044453\Lambda^5 \\ & - 366082940052\Lambda^4 - 649050340979\Lambda^3 - 299118407355\Lambda^2 \\ & - 75936742500\Lambda - 3787700000 \end{aligned} \right) (3C^2 - 1)}{325987320(\Lambda + 1)^3(4\Lambda + 1)(9\Lambda + 1)(9\Lambda + 25)(9\Lambda + 100)(16\Lambda + 1)(81\Lambda + 100)} \right. \\
& + \left. \frac{\left(\begin{aligned} & 73929348\Lambda^7 + 725310531\Lambda^6 + 487357884\Lambda^5 \\ & - 5792477481\Lambda^4 - 9601088132\Lambda^3 - 4427460420\Lambda^2 \\ & - 1076466750\Lambda - 53975000 \end{aligned} \right) (1 - C^2)c_2}{60480(\Lambda + 1)^3(4\Lambda + 1)(9\Lambda + 1)(9\Lambda + 25)(9\Lambda + 100)(16\Lambda + 1)(81\Lambda + 100)} \right\} \lambda\omega \sin 2\omega t \\
& - \left\{ \frac{(1161\Lambda^3 + 14769\Lambda^2 + 74200\Lambda + 40000)(3C^2 - 1)}{221760(\Lambda + 1)^2(4\Lambda + 1)(9\Lambda + 25)(9\Lambda + 100)} \right. \\
& + \left. \frac{(1107\Lambda^3 + 19303\Lambda^2 + 60050\Lambda + 28750)(1 - C^2)c_2}{20160(\Lambda + 1)^2(4\Lambda + 1)(9\Lambda + 25)(9\Lambda + 100)} \right\}
\end{aligned} \tag{61}$$

which cannot be simplified further. In the limit as $\lambda\omega \rightarrow 0$, Eq. (61) reduces to:

$$\psi_4(\theta, \phi, t) = -\frac{1}{945}(3C^2 - 1) - \frac{2323}{30240}(1 - C^2)c_2 \tag{62}$$

which applies to steady shear flow.

VI. RESULTS

We plot a Pipkin space for the misbehavior of each order of the expansion of the orientation distribution function up to ψ_4 . By this, we mean that above the curves in Figure 3 and Figure 4 $\psi < 0$ for at least part of the (θ, ϕ) space over at least part of the cycle ωt .

Keeping terms up to order $(6\lambda\dot{\gamma}^0)^0$ in Eq. (16), we get the orientation distribution for the suspension at rest:

$$\psi(\theta, \phi, t) = \frac{1}{4\pi} \tag{63}$$

and keeping terms up to $(6\lambda\dot{\gamma}^0)^1$ in Eq. (16), we get:

$$\psi(\theta, \phi, t) = \frac{1}{4\pi} \left[1 + (6\lambda\dot{\gamma}^0) \left\{ \frac{1}{12} P_2^2 s_2(1; t) \right\} \right] \quad (64)$$

Substituting Eq. (71) and P_2^2 from Table III into Eq. (64):

$$\psi(\theta, \phi, t) = \frac{1}{4\pi} \left[1 + (6\lambda\dot{\gamma}^0) \left\{ \frac{1}{4} (1 - C^2) s_2 \frac{\cos \omega t + \text{De} \sin \omega t}{1 + L} \right\} \right] \quad (65)$$

which goes negative when:

$$1 + (6\lambda\dot{\gamma}^0) \left\{ \frac{1}{4} (1 - C^2) s_2 \frac{\cos \omega t + \lambda \omega \sin \omega t}{1 + L} \right\} < 0 \quad (66)$$

and since the lowest value of $(1 - C^2) s_2$ is -1 , Eq. (64) predicts negative ψ when:

$$1 - (6\lambda\dot{\gamma}^0) \left\{ \frac{1}{4} \frac{\cos \omega t + \lambda \omega \sin \omega t}{1 + L} \right\} < 0 \quad (67)$$

and since the maximum value of $(\cos \omega t + \text{De} \sin \omega t)$ occurs when $\tan \omega t = \lambda \omega$, Eq. (64) goes negative when:

$$(6\lambda\dot{\gamma}^0) \left\{ \frac{1}{4\sqrt{1+L}} \right\} > 1 \quad (68)$$

or when:

$$\frac{3\lambda\dot{\gamma}^0}{2\sqrt{1+L}} > 1 \quad (69)$$

and taking the limit as $\omega \rightarrow 0$ we see that for steady shear flow, Eq. (64) goes negative when:

$$\lambda\dot{\gamma}^0 > \frac{2}{3} \quad (70)$$

we would normally think of negative ψ as unphysical, though whole papers have been written about ψ in steady shear flow where $\lambda\dot{\gamma}^0 > 2/3$ [31,32], including discussions of the expansion in Eq. (16).

For orders of the expansion above $(6\lambda\dot{\gamma}^0)^1$, we must evaluate $\psi(\theta, \phi, t)$ numerically over the range of $\omega t = [0 : 2\pi]$. We plot these curves and the curve given by Eq. (69) in Figure 3 and Figure 4. We find that interesting behavior occurs when $\lambda\dot{\gamma}^0$ exceeds a certain limiting value (the space in Figure 3 and Figure 4 above each of the curves). Traversing this limit, the orientation distribution function predicts negative $\psi(\theta, \phi, t)$ over four small spots on the (θ, ϕ) space (with only two spots negative at a time, and this for just a short interval of ωt). This misbehavior also arises for steady shear flow ($\lambda\omega \rightarrow 0$), and it has not been observed previously [31,32].

We find that at small enough $\lambda\dot{\gamma}^0$, the farthest term to the right in the diffusion equation becomes negligible and the solution for the orientation distribution function becomes spherical. For cases of constant $\lambda\dot{\gamma}^0$, as the $\lambda\omega$ increases, the distortion from a random orientation is reduced. From this, we see that the rigid dumbbells, in a linearly viscoelastic flow, will remain nearly

isotropic (oriented randomly). The key word here is nearly. In other words, only slight departures from the isotropic state are sufficient to explain, at least qualitatively, the measured behavior in small-amplitude oscillatory shear flow (see figures Fig. 3d. and Fig. 3e. in [23]; see derivation of Eqs. (88) and (89) for η' and η'' in [12]).

Figure 5 through Figure 22 illustrate our results for the expansions of the orientation distribution function, Eq. (16), for all four orders of $(6\lambda\dot{\gamma}^0)$. The color red in these figures indicates the highest magnitudes of ψ , and blue, the lowest. To more clearly illustrate the orientations, each tile in the figures has been normalized. The arc labeled ωt in each figure indicates each of eight angular positions ($0 = \omega t = 2\pi$) during each alternant cycle ($\omega t = 0, \frac{\pi}{4}, \frac{\pi}{2}, \dots, 2\pi$).

We present figures for the truncated expansions given by Eq. (16) up to $(6\lambda\dot{\gamma}^0)^1$, $(6\lambda\dot{\gamma}^0)^2$, $(6\lambda\dot{\gamma}^0)^3$ and $(6\lambda\dot{\gamma}^0)^4$. We remind the reader that each of these expansions uncovers a new layer of the physics of the orientations. For instance, whereas $(6\lambda\dot{\gamma}^0)^1$ just produces orientation at the test frequency ω , $(6\lambda\dot{\gamma}^0)^2$ adds both a time steady component to the orientation as well as orientation 2ω , $(6\lambda\dot{\gamma}^0)^3$ then adds both the third harmonic, and a correction to the ω -component, and then finally, $(6\lambda\dot{\gamma}^0)^4$ introduces the fourth harmonic of the orientation, along with corrections to the time steady component and the 2ω -component. Hence, Figure 5 through Figure 8 explore the $(6\lambda\dot{\gamma}^0)^1$ expansion, Figure 9 through Figure 12 explore $(6\lambda\dot{\gamma}^0)^2$, Figure 13 through Figure 17, $(6\lambda\dot{\gamma}^0)^3$, and Figure 18 through Figure 22, $(6\lambda\dot{\gamma}^0)^4$. We have explored a 3x3 grid on the log-log Pipkin space to cover the widest range of orientation behaviors, and specifically to include Newtonian, linear viscoelastic, nonlinear viscoelastic, and generalized Newtonian flows. Following Figure 3 or Figure 4, we have of course excluded, cases where $\psi < 0$ arises for any part of the (θ, ϕ) space over any part of the cycle ωt .

To help the reader navigate through our results, that is, through Figure 5 to Figure 22, we have constructed Table IV. Using Table IV, we see that to explore the effect of $\lambda\omega$, we can examine Figure 18 through Figure 20 for $\lambda\dot{\gamma}^0 = 0.1$, or Figure 21 and Figure 22 for $\lambda\dot{\gamma}^0 = 1$. In this connection, examining Figure 18 through Figure 20 for $\lambda\dot{\gamma}^0 = 0.1$, we find the orientation distribution become more isotropic with increasing $\lambda\omega$, and we observe the same trend for $\lambda\dot{\gamma}^0 = 1$ (compare Figure 21 with Figure 22). Figure 21 reveals beautiful peanut-shaped orientations distributions over most of the cycle ($\omega t = 0, \frac{\pi}{4}, \frac{\pi}{2}, \pi, \frac{5\pi}{4}, \frac{3\pi}{2}, 2\pi$), but is only slightly oriented at $\omega t = \frac{3\pi}{4}, \frac{7\pi}{4}$. We cannot help but notice that these peanut-shapes resemble the molecular model itself (compare with Figure 2).

Using Table IV, we can also see that to explore the effect of $\lambda\dot{\gamma}^0$, we can examine Figure 19 and Figure 21 for $\lambda\omega = 0.1$, or Figure 20 and Figure 22 for $\lambda\omega = 1$. In this connection, examining Figure 19 and Figure 21 for $\lambda\omega = 0.1$, we see that increasing the dimensionless shear rate, $\lambda\dot{\gamma}^0$, substantially orients the material in oscillatory shear. To a lesser extent we observe the same effect by comparing Figure 20 and Figure 22 for $\lambda\omega = 1$.

To see how the higher harmonics of the orientation shape the orientation distribution at $\lambda\dot{\gamma}^0 = 0.1$, we can examine Figure 5, Figure 9, Figure 13 and Figure 18 for $\lambda\omega = 0.1$, or Figure 6, Figure 10, Figure 14 and Figure 19 for $\lambda\omega = 1$, or Figure 7, Figure 11, Figure 15 and Figure 20 for $\lambda\omega = 10$. In this connection, examining Figure 5, Figure 9, Figure 13 and Figure 18 for $\lambda\omega = 0.1$, we can see little change in the shape of the orientation distribution. This, of course, is because the test conditions of $\lambda\omega = \lambda\dot{\gamma}^0 = 0.1$ correspond to the Newtonian flow regime (near the origin of the Pipkin maps in Figure 3 or Figure 4), where only a little orientation is expected. Examining Figure 6, Figure 10, Figure 14 and Figure 19 for $\lambda\omega = 1$, we also see little change in the shape of the orientation distribution. As discussed earlier in this section, this is because, for linear viscoelasticity, only slight departures from the isotropic state arise. Next, examining Figure 7, Figure 11, Figure 15 and Figure 20 for $\lambda\omega = 10$,

Finally, to see how the higher harmonics of the orientation shape the orientation distribution at $\lambda\dot{\gamma}^0 = 1$, we can examine Figure 16 and Figure 21 for $\lambda\omega = 1$, or Figure 8, Figure 12, Figure 17 and Figure 22 for $\lambda\omega = 10$. Examining Figure 16 and Figure 21 for $\lambda\omega = 1$, or Figure 8, Figure 12, Figure 17 and Figure 22 for $\lambda\omega = 10$, we see little reshaping of the orientation distribution function from the higher harmonics (even for test conditions generating peanut-shaped distributions). In our previous work, deriving the expressions for the shear stress (Eq. (82) of [12]) and for the normal stress differences (Eqs. (13) and (21) of [13]) in large-amplitude oscillatory shear flow, we found that the higher order terms, and thus the higher harmonics, of the orientation distribution function make significant contributions to the rheological properties. From this we learn that even subtle tweaking of the molecular motions, too subtle to observe in our color figures, can matter for the rheology of the polymeric liquid.

VII. CONCLUSION

We have used the dilute suspension of rigid dumbbells in a Newtonian solvent to model the behavior of polymeric liquids in oscillatory shear flow, including especially large-amplitude oscillations. We have solved the diffusion equation [Eq. (6)] analytically to explore the orientations of the molecules induced by the oscillatory shear flow. We find zeroth, first, second, third and fourth harmonics of the orientation distribution function, and we have derived expressions for these [Eq. (16) with Eqs. (36), (44), (52) and (61)].

The orientation distribution function gives information about a single molecule in a sea of solvent. This molecule, under a large-amplitude oscillatory

shearing deformation, is in motion, and occupies a continuous distribution of orientations at any given position in the cycle ωt . This motion is subject to convection [rightmost term in Eq. (6)] and Brownian forces [first term on right of Eq. (6)]. We have used the diffusion equation [Eq. (16) with Eqs. (36), (44), (52) and (61)] to elucidate these molecular motions by rendering the orientation distribution surfaces all the way around each alternant cycle of oscillatory shear. Specifically, Figure 18 through Figure 22 explore the effects of the shear rate, $\lambda\dot{\gamma}^0$, and the test frequency, $\lambda\omega$, on the molecular orientations in oscillatory shear. This exploration includes the Newtonian (Figure 18), the linear viscoelastic (Figure 19 and Figure 20) and nonlinear viscoelastic (Figure 21 and Figure 22) regimes.

A real polymeric liquid, of course, consists of a collection of molecules. So our model for $\psi(\theta, \phi, t)$, also sheds light on the mole fraction of molecules occupying any particular orientation, at any given position in the cycle ωt . The integral of $\psi(\theta, \phi, t)$ over any space (θ_1, θ_2) and (ϕ_1, ϕ_2) thus gives the mole fraction of molecules occupying orientations in that space.

We have used the successive expansions of the orientation distribution function to explore how the addition of each successive harmonic shapes the orientation distribution, and we have explored this over Pipkin space.

We have discovered misbehavior in our analytical solution to the orientation distribution function [Eq. (16) with Eqs. (36), (44), (52) and (61)], and we have delimited the range of shear rates, $\lambda\dot{\gamma}^0$, and the test frequencies, $\lambda\omega$, that produces this misbehavior. We have mapped these ranges out in Pipkin space (see Figure 3 and Figure 4).

Our theory for the viscoelasticity of the polymeric liquid in oscillatory shear relies entirely on the orientation distribution function to explain the shear stress (Eq. (82) of [12]) and the normal stress difference responses (Eqs. (13) and (21) of [13]). Of course the shear stress response causes viscous heating, and thus the temperature to rise [33,34,35]. To explore the role of molecular orientation, and thus molecular structure on the temperature rise in oscillatory shear, we need only follow the method of [35] using Eq. (82) of [12].

VIII. ACKNOWLEDGMENT

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IX. APPENDIX A. FIRST AND SECOND ORDER PAREN FUNCTIONS

For calculating the orientation distribution function, we need:

$$(1;t) \cos \omega t = \frac{1}{2} \frac{\cos 2\omega t + \lambda \omega \sin 2\omega t}{1 + \Lambda} + \frac{1}{2} \left(\frac{1}{1 + \Lambda} \right) \quad (71)$$

$$(0,1;t) = \frac{[1 - 2\Lambda] \cos 2\omega t + 3\lambda \omega \sin 2\omega t}{2(1 + 4\Lambda)(1 + \Lambda)} + \frac{1}{2(1 + \Lambda)} \quad (72)$$

$$\left(\frac{7}{3},1;t\right) = \frac{3(10 - 6\Lambda) \cos 2\omega t + 48\lambda \omega \sin 2\omega t}{16(25 + 9\Lambda)(1 + \Lambda)} + \frac{3}{20(1 + \Lambda)} \quad (73)$$

in which $\Lambda = (\lambda \omega)^2$.

X. APPENDIX B. THE THIRD ORDER PAREN FUNCTIONS

For any values of a , b , and c we have (see Eq. (71) in [12]):

$$(a,b,c;t) = \left[\begin{aligned} & \frac{\left(\left[(a+b+c)(b+c)c - \{2(a+b+c) + 3(b+c) + 6c\} \Lambda \right] \cos 3\omega t \right.}{4 \left((a+b+c)^2 + 9\Lambda \right) \left((b+c)^2 + 4\Lambda \right) (c^2 + \Lambda)} \\ & \left. + \frac{\left[-6\Lambda + \{ (a+b+c)(b+c) + 2(a+b+c)c + 3(b+c)c \} \right] \lambda \omega \sin 3\omega t}{4 \left((a+b+c)^2 + 9\Lambda \right) \left((b+c)^2 + 4\Lambda \right) (c^2 + \Lambda)} \right) \\ & + \frac{\left(\left[(a+b+c)(b+c)c - \{2(a+b+c) + (b+c) + 2c\} \Lambda \right] \cos \omega t \right.}{4 \left((a+b+c)^2 + \Lambda \right) \left((b+c)^2 + 4\Lambda \right) (c^2 + \Lambda)} \\ & \left. + \frac{\left[-2\Lambda + \{ (a+b+c)(b+c) + 2(a+b+c)c + (b+c)c \} \right] \lambda \omega \sin \omega t}{4 \left((a+b+c)^2 + \Lambda \right) \left((b+c)^2 + 4\Lambda \right) (c^2 + \Lambda)} \right) \\ & + 2 \frac{\left[(a+b+c)c \right] \cos \omega t + (c)(\lambda \omega) \sin \omega t}{4 \left((a+b+c)^2 + \Lambda \right) (b+c)(c^2 + \Lambda)} \end{aligned} \right] \quad (74)$$

or,

$$[A,B,1;t] = \left[\begin{aligned} & \frac{\left(\left[AB - (2A + 3B + 6)\Lambda \right] \cos 3\omega t \right.}{4 \left(A^2 + 9\Lambda \right) \left(B^2 + 4\Lambda \right) (1 + \Lambda)} \\ & \left. + \frac{\left[-6\Lambda + (AB + 2A + 3B) \right] \lambda \omega \sin 3\omega t}{4 \left(A^2 + 9\Lambda \right) \left(B^2 + 4\Lambda \right) (1 + \Lambda)} \right) \\ & + \frac{\left(\left[(3AB^2 + 8A\Lambda) - (2AB + B^2 + 2B)\Lambda \right] \cos \omega t \right.}{4 \left(A^2 + \Lambda \right) B \left(B^2 + 4\Lambda \right) (1 + \Lambda)} \\ & \left. + \frac{\left[(AB^2 + 2AB + 3B^2) + (8 - 2B)\Lambda \right] \lambda \omega \sin \omega t}{4 \left(A^2 + \Lambda \right) B \left(B^2 + 4\Lambda \right) (1 + \Lambda)} \right) \end{aligned} \right] \quad (75)$$

where the “bracket functions” are given in terms of $A \equiv a + b + c$, $B \equiv b + c$, and $C = c = 1$ (the only case for c that is needed). The five specific cases that are necessary for the evaluation of the orientation distribution function are:

$$(0,0,1;t)=[1,1,1;t]=\left[\begin{array}{l}\frac{[1-11\Lambda]\cos 3\omega t+6[1-\Lambda]\lambda\omega\sin 3\omega t}{4(1+9\Lambda)(1+4\Lambda)(1+\Lambda)}\\+\frac{3\cos \omega t+6\lambda\omega\sin \omega t}{4(1+4\Lambda)(1+\Lambda)}\end{array}\right] \quad (76)$$

$$\left(-\frac{7}{3},\frac{7}{3},1;t\right)=\left[1,\frac{10}{3},1;t\right]=\left[\begin{array}{l}\frac{3(5-27\Lambda)\cos 3\omega t+3(23-9\Lambda)\lambda\omega\sin 3\omega t}{8(1+9\Lambda)(25+9\Lambda)(1+\Lambda)}\\+\frac{3(75-37\Lambda)\cos \omega t+3(3\Lambda+115)\lambda\omega\sin \omega t}{40(25+9\Lambda)(1+\Lambda)^2}\end{array}\right] \quad (77)$$

$$\left(\frac{7}{3},0,1;t\right)=\left[\frac{10}{3},1,1;t\right]=\left[\begin{array}{l}\frac{3(10-47\Lambda)\cos 3\omega t+9(13-6\Lambda)\lambda\omega\sin 3\omega t}{4(1+4\Lambda)(100+81\Lambda)(1+\Lambda)}\\+\frac{9(10+17\Lambda)\cos \omega t+9(13+6\Lambda)\lambda\omega\sin \omega t}{4(1+4\Lambda)(100+9\Lambda)(1+\Lambda)}\end{array}\right] \quad (78)$$

$$\left(0,\frac{7}{3},1;t\right)=\left[\frac{10}{3},\frac{10}{3},1;t\right]=\left[\begin{array}{l}\frac{18(25-51\Lambda)\cos 3\omega t+9(125-27\Lambda)\lambda\omega\sin 3\omega t}{8(25+9\Lambda)(100+81\Lambda)(1+\Lambda)}\\+\frac{270(25-3\Lambda)\cos \omega t+9(625+9\Lambda)\lambda\omega\sin \omega t}{40(1+4\Lambda)(100+9\Lambda)(1+\Lambda)}\end{array}\right] \quad (79)$$

$$\left(\frac{11}{3},\frac{7}{3},1;t\right)=\left[\frac{21}{3},\frac{10}{3},1;t\right]=\left[\begin{array}{l}\frac{15(7-9\Lambda)\cos 3\omega t+3(71-9\Lambda)\lambda\omega\sin 3\omega t}{8(25+9\Lambda)(49+9\Lambda)(1+\Lambda)}\\+\frac{3(525-19\Lambda)\cos \omega t+3(355+3\Lambda)\lambda\omega\sin \omega t}{40(25+9\Lambda)(1+\Lambda)(49+\Lambda)}\end{array}\right] \quad (80)$$

XI. APPENDIX C. THE FOURTH ORDER PAREN FUNCTIONS $(a,b,c,d;t)$

For any values of a , b , c , and d we have (see Eq. (48) in [13,14]):

$$\begin{aligned}
(a, b, c, d; t) = & \frac{\left\{ \begin{aligned} & \left[\left[(a+b+c+d)(b+c+d) - 12\Lambda \right] \left[(c+d)d - 2\Lambda \right] \right\} \cos 4\omega t \\ & - \left[3(a+b+c+d) + 4(b+c+d) \right] \left[(c+d) + 2d \right] \Lambda \end{aligned} \right\}}{8 \left((a+b+c+d)^2 + 16\Lambda \right) \left((b+c+d)^2 + 9\Lambda \right) \left((c+d)^2 + 4\Lambda \right) (d^2 + \Lambda)} \\
& + \frac{\left\{ \begin{aligned} & \left[\left[(a+b+c+d)(b+c+d) - 12\Lambda \right] \left[(c+d) + 2d \right] \right\} \lambda \omega \sin 4\omega t \\ & + \left[3(a+b+c+d) + 4(b+c+d) \right] \left[(c+d)d - 2\Lambda \right] \end{aligned} \right\}}{8 \left((a+b+c+d)^2 + 16\Lambda \right) \left((b+c+d)^2 + 9\Lambda \right) \left((c+d)^2 + 4\Lambda \right) (d^2 + \Lambda)} \\
& + \frac{\left\{ \begin{aligned} & \left[\left[(a+b+c+d)(b+c+d) - 6\Lambda \right] \left[(c+d)d - 2\Lambda \right] \right\} \cos 2\omega t \\ & - \left[3(a+b+c+d) + 2(b+c+d) \right] \left[(c+d) + 2d \right] \Lambda \end{aligned} \right\}}{8 \left((a+b+c+d)^2 + 4\Lambda \right) \left((b+c+d)^2 + 9\Lambda \right) \left((c+d)^2 + 4\Lambda \right) (d^2 + \Lambda)} \\
& + \frac{\left\{ \begin{aligned} & \left[\left[(a+b+c+d)(b+c+d) - 6\Lambda \right] \left[(c+d) + 2d \right] \right\} \lambda \omega \sin 2\omega t \\ & + \left[3(a+b+c+d) + 2(b+c+d) \right] \left[(c+d)d - 2\Lambda \right] \end{aligned} \right\}}{8 \left((a+b+c+d)^2 + 4\Lambda \right) \left((b+c+d)^2 + 9\Lambda \right) \left((c+d)^2 + 4\Lambda \right) (d^2 + \Lambda)} \\
& + \frac{\left\{ \begin{aligned} & \left[\left[(a+b+c+d)(b+c+d) - 2\Lambda \right] \left[(c+d)d - 2\Lambda \right] \right\} \cos 2\omega t \\ & - \left[(a+b+c+d) + 2(b+c+d) \right] \left[(c+d) + 2d \right] \Lambda \end{aligned} \right\}}{8 \left((a+b+c+d)^2 + 4\Lambda \right) \left((b+c+d)^2 + \Lambda \right) \left((c+d)^2 + 4\Lambda \right) (d^2 + \Lambda)} \\
& + \frac{\left\{ \begin{aligned} & \left[\left[(a+b+c+d)(b+c+d) - 2\Lambda \right] \left[(c+d) + 2d \right] \right\} \lambda \omega \sin 2\omega t \\ & + \left[(a+b+c+d) + 2(b+c+d) \right] \left[(c+d)d - 2\Lambda \right] \end{aligned} \right\}}{8 \left((a+b+c+d)^2 + 4\Lambda \right) \left((b+c+d)^2 + \Lambda \right) \left((c+d)^2 + 4\Lambda \right) (d^2 + \Lambda)} \\
& + \frac{\left\{ \begin{aligned} & \left[\left[(a+b+c+d)(b+c+d) - 2\Lambda \right] d - \left[(a+b+c+d) + 2(b+c+d) \right] \Lambda \right] \cos 2\omega t \\ & + \left[\left[(a+b+c+d)(b+c+d) - 2\Lambda \right] + \left[(a+b+c+d) + 2(b+c+d) \right] d \right] \lambda \omega \sin 2\omega t \end{aligned} \right\}}{8 \left((a+b+c+d)^2 + \Lambda \right) \left((b+c+d)^2 + \Lambda \right) (c+d) (d^2 + \Lambda)} \\
& + \frac{\left\{ \begin{aligned} & \left[\left[(a+b+c+d)(b+c+d) - 2\Lambda \right] d + \left[(a+b+c+d) + 2(b+c+d) \right] \Lambda \right] \cos 2\omega t \\ & - \left[\left[(a+b+c+d)(b+c+d) - 2\Lambda \right] - \left[(a+b+c+d) + 2(b+c+d) \right] d \right] \lambda \omega \sin 2\omega t \end{aligned} \right\}}{8 \left((a+b+c+d)^2 + 4\Lambda \right) \left((b+c+d)^2 + \Lambda \right) (c+d) (d^2 + \Lambda)} \\
& + \frac{\left[(b+c+d)(c+d) - 2\Lambda \right] d - \left[2(b+c+d) + (c+d) \right] \Lambda}{8 \left((a+b+c+d) \left((b+c+d)^2 + \Lambda \right) \left((c+d)^2 + 4\Lambda \right) (d^2 + \Lambda) \right)} \\
& + \frac{2(b+c+d)d}{8 \left((a+b+c+d) \left((b+c+d)^2 + \Lambda \right) (c+d) (d^2 + \Lambda) \right)}
\end{aligned} \tag{81}$$

For any values of a , b , c , and d (but with $d=1$, being the only case of interest here), and with, $A \equiv a+b+c+d$, $B \equiv b+c+d$, $C \equiv c+d$ and $D \equiv d=1$ we have:

$$\begin{aligned}
(a, b, c, 1; t) = [A, B, C, 1; t] = & \frac{[(B-12\Lambda)(C-2\Lambda)-(3+4B)(C+2)\Lambda]\cos 4\omega t}{8(1+16\Lambda)(B^2+9\Lambda)(C^2+4\Lambda)(1+\Lambda)} \\
& + \frac{\left[\begin{aligned} & [(B-6\Lambda)(C-2\Lambda)-(3+2B)(C+2)\Lambda](B^2+\Lambda)(1+\Lambda)C \\ & + [(B-2\Lambda)(C-2\Lambda)-(1+2B)(C+2)\Lambda](B^2+9\Lambda)(1+\Lambda)C \\ & + [(B-2\Lambda)-(1+2B)\Lambda](1+4\Lambda)(B^2+9\Lambda)(C^2+4\Lambda) \\ & + [(B-2\Lambda)+(1+2B)\Lambda](B^2+9\Lambda)(C^2+4\Lambda)(1+\Lambda) \end{aligned} \right]}{8(1+4\Lambda)(B^2+9\Lambda)(C^2+4\Lambda)(1+\Lambda)^2(B^2+\Lambda)C} \cos 2\omega t \\
& + \frac{\left[\begin{aligned} & [(B-6\Lambda)(C+2)+(3+2B)(C-2\Lambda)](B^2+\Lambda)(1+\Lambda)C \\ & + [(B-2\Lambda)(C+2)+(1+2B)(C-2\Lambda)](B^2+9\Lambda)(1+\Lambda)C \\ & + [(B-2\Lambda)+(1+2B)](1+4\Lambda)(B^2+9\Lambda)(C^2+4\Lambda) \\ & + [(1+2B)-(B-2\Lambda)](B^2+9\Lambda)(C^2+4\Lambda)(1+\Lambda) \end{aligned} \right]}{8(1+4\Lambda)(B^2+9\Lambda)(C^2+4\Lambda)(1+\Lambda)^2(B^2+\Lambda)C} \lambda\omega \sin 2\omega t \\
& + \frac{3BC^2 + (8B-2C-2BC-C^2)\Lambda}{8(B^2+\Lambda)(C^2+4\Lambda)(1+\Lambda)C}
\end{aligned} \tag{82}$$

Next we evaluate the $(a, b, c, 1; t) = [A, B, C, 1; t]$ for the cases we need for getting the orientation distribution function:

$$(0, 0, 0, 1; t) = [1, 1, 1, 1; t] = \left[\begin{aligned} & \frac{1}{8} \frac{(1-35\Lambda+24\Lambda^2)\cos 4\omega t + (10-50\Lambda)\lambda\omega \sin 4\omega t}{(1+16\Lambda)(1+9\Lambda)(1+4\Lambda)(1+\Lambda)} \\ & + \frac{1}{8} \frac{(4-13\Lambda-32\Lambda^2-159\Lambda^3)\cos 2\omega t + 2(10+26\Lambda+61\Lambda^2-27\Lambda^3)\lambda\omega \sin 2\omega t}{(1+4\Lambda)(1+9\Lambda)(1+\Lambda)^3} \\ & + \frac{3}{8} \frac{1}{(1+4\Lambda)(1+\Lambda)} \end{aligned} \right] \tag{83}$$

$$(0, -\frac{7}{3}, \frac{7}{3}, 1; t) = [1, 1, \frac{10}{3}, 1; t] = \left[\begin{aligned} & \frac{3}{4} \frac{(5-119\Lambda+36\Lambda^2)\cos 4\omega t + (43-117\Lambda)\lambda\omega \sin 4\omega t}{(1+16\Lambda)(1+9\Lambda)(100+36\Lambda)(1+\Lambda)} \\ & + \frac{3}{20} \frac{\left[(100+243\Lambda-2289\Lambda^2-5915\Lambda^3-1179\Lambda^4)\cos 2\omega t + 2(215+1447\Lambda+1934\Lambda^2-693\Lambda^3-243\Lambda^4)\lambda\omega \sin 2\omega t \right]}{(1+4\Lambda)(1+9\Lambda)(100+36\Lambda)(1+\Lambda)^3} \\ & + \frac{3}{20} \frac{(75-37\Lambda)}{(1+\Lambda)^2(100+36\Lambda)} \end{aligned} \right] \tag{84}$$

$$\left(-\frac{7}{3}, \frac{7}{3}, 0, 1; t\right) = \left[1, \frac{10}{3}, 1, 1; t\right] = \left[\begin{aligned} & \frac{3}{8} \frac{(10 - 203\Lambda + 72\Lambda^2) \cos 4\omega t + (79 - 206\Lambda) \lambda \omega \sin 4\omega t}{(1 + 16\Lambda)(100 + 81\Lambda)(1 + 4\Lambda)(1 + \Lambda)} \\ & + \frac{3}{8} \frac{\left[(4000 - 5680\Lambda - 10262\Lambda^2 - 38031\Lambda^3 - 30780\Lambda^4) \cos 2\omega t \right.}{(1 + 16\Lambda)(100 + 81\Lambda)(1 + 4\Lambda)(1 + \Lambda)^2(100 + 9\Lambda)} \\ & \left. + (15800 + 35050\Lambda + 63881\Lambda^2 + 32130\Lambda^3 - 5832\Lambda^4) \lambda \omega \sin 2\omega t \right] \end{aligned} \right] \quad (85)$$

$$\left(-\frac{7}{3}, 0, \frac{7}{3}, 1; t\right) = \left[1, \frac{10}{3}, \frac{10}{3}, 1; t\right] = \left[\begin{aligned} & \frac{9}{4} \frac{2(25 - 301\Lambda + 54\Lambda^2) \cos 4\omega t + 5(65 - 87\Lambda) \lambda \omega \sin 4\omega t}{(1 + 16\Lambda)(100 + 81\Lambda)(100 + 36\Lambda)(1 + \Lambda)} \\ & + \frac{9}{20} \frac{\left[(100000 - 72000\Lambda - 475930\Lambda^2 - 330813\Lambda^3 - 62451\Lambda^4) \cos 2\omega t \right.}{(1 + 4\Lambda)(100 + 81\Lambda)(100 + 36\Lambda)(1 + \Lambda)^2(100 + 9\Lambda)} \\ & \left. + (325000 + 622150\Lambda + 269979\Lambda^2 - 4725\Lambda^3 - 13122\Lambda^4) \lambda \omega \sin 2\omega t \right] \\ & + \frac{27}{2} \frac{(25 - 3\Lambda)}{(100 + 9\Lambda)(100 + 36\Lambda)(1 + \Lambda)} \end{aligned} \right] \quad (86)$$

Table I: Dimensional Variables

Velocity, <i>i</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 1)	L	y
Cartesian coordinate, flow direction (Figure 1)	L	x
Cartesian coordinate, transverse to flow direction (Figure 1)	L	z
Gap	L	h
Molar concentration of dumbbells	$\text{dumbbells}/L^3$	n
Past time	t	t', t'', t'''
Relaxation time of fluid	t	λ
Shear rate amplitude	t^{-1}	$\dot{\gamma}^0$
Solvent viscosity	M/Lt	η_s
Temperature	T	T
Time	t	t

Legend: M = mass; L = length; t = time; T = temperature

**See Eqs. (4.4-14) and (4.4-15) and also NOTATION FOR VOLUME 1: Greek Symbols of [36].

Table II: Dimensionless Variables and Groups

Azimuthal angle	ϕ
Deborah number squared	$\Lambda \equiv (\lambda\omega)^2$
Deborah number, oscillatory shear	$\lambda\omega$
Number of equally-spaced beads on a rod of length $(L - b)$	N
Orientation distribution	$\psi(\theta, \phi, t)$
Paren function, first order	$(a; t)$
Paren function, fourth order	$(a, b, c, d; t)$
Paren function, second order	$(a, b; t)$
Paren function, third order	$(a, b, c; t)$
Polar angle	θ
Time dependent part of shear rate for velocity profile, assumed linear in y [Eq. (1)]	$f(t)$
Weissenberg number, oscillatory shear	$\lambda\dot{\gamma}^0$

Table III: The Associated Legendre Polynomials P_n^m in Terms of $C \equiv \cos \theta$

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

$$P_4^0(C) = \frac{1}{8}(35C^4 - 30C^2 + 3)$$
$$P_4^2(C) = \frac{15}{2}(1 - C^2)(7C^2 - 1)$$
$$P_4^4(C) = 105(1 - C^2)^2$$

$$P_6^2(C) = \frac{105}{8}(1 - C^2)(33C^4 - 18C^2 + 1)$$
$$P_6^6(C) = 10395(1 - C^2)^3$$

Table IV: Test Conditions and Expansion Order for Results

	Order of Expansion	$\lambda\omega$	$\lambda\dot{\gamma}^0$
Figure 5	$\psi(\psi_1)$	0.1	0.1
Figure 6		1	
Figure 7		10	
Figure 8		10	1
Figure 9	$\psi(\psi_1, \psi_2)$	0.1	0.1
Figure 10		1	
Figure 11		10	
Figure 12		10	1
Figure 13	$\psi(\psi_1, \psi_2, \psi_3)$	0.1	0.1
Figure 14		1	
Figure 15		10	
Figure 16		1	1
Figure 17		10	
Figure 18	$\psi(\psi_1, \psi_2, \psi_3, \psi_4)$	0.1	0.1
Figure 19		1	
Figure 20		10	
Figure 21		1	1
Figure 22		10

Figure 1: Orthomorphic sketch of alternating velocity profile in oscillatory shear between stationary and moving plates [Eqs. (1) or (2)]. Cartesian coordinates with origin on the stationary plate.

Figure 2: The dumbbell consists of identical beads (of identical mass and of identical diameter) connected by one thin, absolutely rigid (dimensionless, massless) rod.

Figure 3: Pipkin space of the orientation distribution function $\psi(\theta, \phi, t)$, for increasing the order of the expansion of $\psi(\theta, \phi, t)$. Above each curve for at least part of the (θ, ϕ) space over at least part of the cycle ωt .

Figure 4: Pipkin space of the orientation distribution function $\psi(\theta, \phi, t)$, for increasing the order of the expansion of $\psi(\theta, \phi, t)$. Above each curve for at least part of the (θ, ϕ) space over at least part of the cycle ωt .

Figure 5: $\psi(\psi_1)$ from Eq. (36), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 0.1$ and $\lambda\dot{\gamma}^0 = 0.1$. Only the first harmonic contributes to these orientations. Newtonian regime.

Figure 6: $\psi(\psi_1)$ from Eq. (36), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 0.1$. Only the first harmonic contributes to these orientations. Linear viscoelastic regime.

Figure 7: $\psi(\psi_1)$ from Eq. (36), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 10$ and $\lambda\dot{\gamma}^0 = 0.1$. Only the first harmonic contributes to these orientations. Linear viscoelastic regime.

Figure 8: $\psi(\psi_1)$ from Eq. (36), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 10$ and $\lambda\dot{\gamma}^0 = 1$. Only the first harmonic contributes to these orientations. Nonlinear viscoelastic regime.

Figure 9: $\psi(\psi_1, \psi_2)$ from Eqs. (36) and (44), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 0.1$ and $\lambda\dot{\gamma}^0 = 0.1$. Only the zeroth, first and second harmonics contribute to these orientations. Newtonian regime.

Figure 10: $\psi(\psi_1, \psi_2)$ from Eqs. (36) and (44), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 0.1$. Only the zeroth, first and second harmonics contribute to these orientations. Linear viscoelastic regime.

Figure 11: $\psi(\psi_1, \psi_2)$ from Eqs. (36) and (44), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 10$ and $\lambda\dot{\gamma}^0 = 0.1$. Only the zeroth, first and second harmonics contribute to these orientations. Linear viscoelastic regime.

Figure 12: $\psi(\psi_1, \psi_2)$ from Eqs. (36) and (44), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 10$ and $\lambda\dot{\gamma}^0 = 1$. Only the zeroth, first and second harmonics contribute to these orientations. Nonlinear viscoelastic regime.

Figure 13: $\psi(\psi_1, \psi_2, \psi_3)$ from Eqs. (36), (44), and (52), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 0.1$ and $\lambda\dot{\gamma}^0 = 0.1$. Only the zeroth, first, second, and third harmonics contribute to these orientations. Newtonian regime.

Figure 14: $\psi(\psi_1, \psi_2, \psi_3)$ from Eqs. (36), (44), and (52), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 0.1$. Only the zeroth, first, second, and third harmonics contribute to these orientations. Linear viscoelastic regime.

Figure 15: $\psi(\psi_1, \psi_2, \psi_3)$ from Eqs. (36), (44), and (52), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 10$ and $\lambda\dot{\gamma}^0 = 0.1$. Only the zeroth, first, second, and third harmonics contribute to these orientations. Linear viscoelastic regime.

Figure 16: $\psi(\psi_1, \psi_2, \psi_3)$ from Eqs. (36), (44), and (52), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 1$. Only the zeroth, first, second, and third harmonics contribute to these orientations. Nonlinear viscoelastic regime.

Figure 17: $\psi(\psi_1, \psi_2, \psi_3)$ from Eqs. (36), (44), and (52), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 10$ and $\lambda\dot{\gamma}^0 = 1$. Only the zeroth, first, second, and third harmonics contribute to these orientations. Nonlinear viscoelastic regime.

Figure 18: $\psi(\psi_1, \psi_2, \psi_3, \psi_4)$ from Eqs. (36), (44), (52) and (61), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 0.1$ and $\lambda\dot{\gamma}^0 = 0.1$. Here the zeroth, first, second, third and fourth harmonics contribute to these orientations. Newtonian regime.

Figure 19: $\psi(\psi_1, \psi_2, \psi_3, \psi_4)$ from Eqs. (36), (44), (52) and (61), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 0.1$. Here the zeroth, first, second, third and fourth harmonics contribute to these orientations. Linear viscoelastic regime.

Figure 20: $\psi(\psi_1, \psi_2, \psi_3, \psi_4)$ from Eqs. (36), (44), (52) and (61), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 10$ and $\lambda\dot{\gamma}^0 = 0.1$. Here the zeroth, first, second, third and fourth harmonics contribute to these orientations. Linear viscoelastic regime.

Figure 21: $\psi(\psi_1, \psi_2, \psi_3, \psi_4)$ from Eqs. (36), (44), (52) and (61), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 1$ and $\lambda\dot{\gamma}^0 = 1$. Here the zeroth, first, second, third and fourth harmonics contribute to these orientations. Nonlinear viscoelastic regime.

Figure 22: $\psi(\psi_1, \psi_2, \psi_3, \psi_4)$ from Eqs. (36), (44), (52) and (61), over a complete alternant cycle of $\omega t = 0$ to $\omega t = 2\pi$, with $\lambda\omega = 10$ and $\lambda\dot{\gamma}^0 = 1$. Here the zeroth, first, second, third and fourth harmonics contribute to these orientations. Nonlinear viscoelastic regime.

XII. REFERENCES

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

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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 $L + d$ ".
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