1. INTRODUCTION

Since its conception\(^1\text{-}^3\), and then the perfection of the measurement of both parts of the complex viscosity\(^4\), small shear rate amplitude oscillatory shear flow has become the most popular experiment for investigating the physics of fluid elasticity. When performed at large shear rate amplitude, the shear stress responds as a set of harmonics, at odd-multiples of the test frequency. For any given oscillatory shear experiment, the higher the harmonic frequency, the lower its amplitude. Measurements of these higher harmonics have been challenging, and only recently have such measurements become possible on commercial instrumentation. Specifically, the recent advent of plate-partitioning\(^5\text{-}^8\) circumvents the offending effects of edge distortion and fracture.\(^9\text{-}^{15}\) Analytical solutions for macromolecular models have deepened our understanding of the higher harmonics of the shear stress, and Table I classifies this literature. From the fifth column of Table I, however, we see that the literature is still without a molecular explanation of harmonics beyond the third. This paper is the first to provide an explanation of the molecular origins of the fifth harmonic. By molecular origins, we mean how the macromolecular orientations cause the fifth shear stress harmonic, and how this harmonic is connected to the first and third through macromolecular orientations.

Large-amplitude oscillatory shear (LAOS) flow has also been useful in the rheology laboratory. Confinement of an elastic liquid between sliding plates is a common design element (called a viscous damper) used to damp vibration in machinery\(^16\text{-}^{19}\) (Section 3.3 of Ref. 20; Section 8.7 of Ref. 21; Problem 5.5 of Ref. 22-23). The part of the first harmonic that is in-phase with the vibration provides the viscous dissipation, and thus the damping. However, the higher harmonics of the shear stress response themselves create vibrations at odd multiples of the damped vibration frequency. These parasitic vibrations must also then be damped, and in the best designs, the machinery resonant frequencies must not coincide with these odd multiples of the fundamental vibration frequency.

Large-amplitude oscillatory shear (LAOS) flow has also
been (approximately) generated by trapping fluid between solid sheets passed over and under through a series of calendar rolls (see Figure 2 of Ref. 24). This method is used to induce crystallization leading to ordering in nanoparticle-filled polymers (see Figures 6 and 7 of Ref. 24). One particular application allows material opalescence to be controlled in this way \(^{(20)}\) (see Figure 8 of Ref. 25).

In this paper, we seek a molecular understanding of the origins of the fifth harmonic, and for this the rigid dumbbell presents the simplest model for macromolecular fluids. This rigid dumbbell describes the molecular structure of the polymer with two identical spherical beads joined by a massless fixed rod and this dumbbell is then suspended in a Newtonian solvent (see Figure 1). We consider the simplest case where the bead separation is sufficient for the hydrodynamic interaction to vanish. For continuum models, we know of just one equation that has yielded expressions for the fifth harmonic \(^{(27)}\) (see Table I of Ref. 26,51; see also Table I of Ref. 72) and, in this paper, we will compare our molecular modeling results with this continuum prediction for the corotational Maxwell model (see Eq. (58) of Ref. 51; see also Eq. (62) of Ref. 28).

Our previous work evaluated the shear stress response for the rigid dumbbell suspension in large-amplitude oscillatory shear flow up to and including the third harmonic (Eq. (82) of Ref. 72). We achieved this by deriving the first three terms of the deviation of the orientational distribution function from equilibrium, and we then used these terms to find the averages in the Giesekus expression for the stress tensor. Finally, we identified the terms contributing to the shear stress, and then derived the corresponding paren functions. Our use of the Giesekus expression for the stress tensor connects the macromolecular orientations with the shear stress response, and thus, this step deepens our understanding of the molecular

Table I. Literature on analytical solutions for molecular models in LAOS.

<table>
<thead>
<tr>
<th>Model</th>
<th>Shear Stress Harmonic</th>
<th>First</th>
<th>Third</th>
<th>Fifth</th>
<th>Normal Eq. of [31]</th>
<th>[Ref.]</th>
<th>Corrects n</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kirkwood and Posek (1956, 1967); Posek (1957)</td>
<td>RD,SK</td>
<td>n</td>
<td></td>
<td></td>
<td>(157)</td>
<td>[52,53,54]</td>
<td></td>
</tr>
<tr>
<td>Lodge (1961, 1964)</td>
<td>L</td>
<td>ℓ</td>
<td>0</td>
<td>0</td>
<td></td>
<td>[55,56]</td>
<td></td>
</tr>
<tr>
<td>Paul (1969); Paul (1970); Bharadwaj (2012); Bharadwaj (2015)</td>
<td>RD,SK</td>
<td>n</td>
<td>X</td>
<td></td>
<td>(157)</td>
<td>[57,58,59,60, (52,54)</td>
<td></td>
</tr>
<tr>
<td>Paul and Mazo (1969), Paul (1970)</td>
<td>RR</td>
<td>n</td>
<td>X</td>
<td></td>
<td>(157)</td>
<td>[61,58]</td>
<td></td>
</tr>
<tr>
<td>Bird, Warner and Evans (1971)</td>
<td>RD</td>
<td>n</td>
<td></td>
<td></td>
<td>(157)</td>
<td>[62]</td>
<td></td>
</tr>
<tr>
<td>Bird et al. (1977)</td>
<td>BHS</td>
<td>ℓ</td>
<td>0</td>
<td>0</td>
<td></td>
<td>Table 11.4-2  [63]</td>
<td></td>
</tr>
<tr>
<td>Mou and Mazo (1977)</td>
<td>RR</td>
<td></td>
<td></td>
<td></td>
<td>(183)</td>
<td>[64,61]</td>
<td></td>
</tr>
<tr>
<td>Pearson and Rochefort (1982); Helfand and Pearson (1982)</td>
<td>R</td>
<td>n</td>
<td>X</td>
<td></td>
<td>(147)</td>
<td>[65,66]</td>
<td></td>
</tr>
<tr>
<td>Fan and Bird (1984)</td>
<td>CB</td>
<td>n</td>
<td>X</td>
<td></td>
<td>(157)</td>
<td>[67,65]</td>
<td></td>
</tr>
<tr>
<td>Hoyle (2010)</td>
<td>PP</td>
<td>n</td>
<td>X</td>
<td></td>
<td>(147)</td>
<td>[68]</td>
<td></td>
</tr>
<tr>
<td>Wagner et al. (2011)</td>
<td>R</td>
<td>n</td>
<td>X</td>
<td></td>
<td>(147)</td>
<td>[69]</td>
<td></td>
</tr>
<tr>
<td>Gurnon and Wagner (2012)</td>
<td>G</td>
<td>ℓ</td>
<td>X</td>
<td></td>
<td>(147)</td>
<td>[70]</td>
<td></td>
</tr>
<tr>
<td>Abbasi et al. (2013)</td>
<td>MSF</td>
<td>ℓ</td>
<td>X</td>
<td></td>
<td>(147)</td>
<td>[71]</td>
<td></td>
</tr>
<tr>
<td>Bird et al. (2014)</td>
<td>RD</td>
<td>n</td>
<td>X</td>
<td></td>
<td>(157)</td>
<td>[72]</td>
<td></td>
</tr>
<tr>
<td>Schmalzer et al. (2014)</td>
<td>RD</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>[73,74,75,76]</td>
<td></td>
</tr>
<tr>
<td>Bozorgi et al. (2014); Bozorgi and Underhill (2014)</td>
<td>AS</td>
<td>n</td>
<td>X</td>
<td></td>
<td>(147)</td>
<td>[77,78]</td>
<td></td>
</tr>
<tr>
<td>Giacomin et al. (2014, 2015); Gilbert and Giacomin (2016)</td>
<td>RD</td>
<td>n</td>
<td>X</td>
<td></td>
<td>(157)</td>
<td>[79,80,81]</td>
<td></td>
</tr>
<tr>
<td>This Paper</td>
<td>RD</td>
<td>n</td>
<td>X</td>
<td></td>
<td>(157)</td>
<td>[32]</td>
<td></td>
</tr>
</tbody>
</table>

Legend: AS ≡ active rod suspension; BHS ≡ Bond-Hookean spring; CB ≡ Curtiss-Bird; G ≡ Giesekus; L ≡ Lodge rubberlike liquid; MSF ≡ molecular stress function; PP ≡ pompom; RD ≡ rigid dumbbell; R ≡ reptation; RR ≡ planar rigid ring; SK ≡ shish-kebab; \( N_1, N_2 \) = first and second normal stress differences; \( n = \eta \alpha \gamma \_0 \); \( \ell = \eta \alpha \gamma \_0 \); \(^{(20)}\) = multiple relaxation times.
origins of the fifth harmonic.

Below, we derive the response of the next higher harmonic, the fifth. We achieve this by using an additional term in the computations of the averages in the Giesekus expression for the stress tensor, the fourth order term of the orientational distribution function (Eq. (163) of Ref. 74). We then simplify by carefully identifying (and then dropping) the parts of the molecular orientation that make no contribution to the shear stress.

To deepen our understanding of our main result, we follow Bird et al. 72 by comparing the three new \((\lambda^2)_n\) coefficients in Eq. (20) with corresponding coefficients of the corotational Maxwell model. In our previous work 72, we found a remarkable qualitative likeness between corresponding \((\lambda^2)_n\) coefficients (see Figures 7 and 8 of Ref. 72). By qualitative likeness, we mean that the curves have the same shapes, and that they approach the same asymptotes.

## 2. THEORY

### 2.1 Simple Shear Flow

We begin with the kinematics for oscillatory shear flow:

\[
v_x(y,t) = \left[\dot{y}^0 f(t)\right] y = \left(\dot{y}^0 \cos \omega t\right) y;
\]

\[
v_y = v_z = 0
\]

where \(\dot{y}^0\) is the magnitude of the shear rate, and \(x, y, \) and \(z\) are defined in Figure 2. 29,30 We nondimensionalize Eq. (1) with the characteristic relaxation time for the fluid, \(\lambda\) (see Eqs. (6) and (7) of Ref. 72):

\[
v_x(y,t) = \left[\lambda \dot{y}^0 \cos (\lambda \omega (t / \lambda))\right] (y / \lambda)
\]

where \(\lambda \dot{y}^0\) and \(\lambda \omega\) are called the Weissenberg and the Deborah numbers. Our symbols are defined, along with their dimensions, in Table II and Table III.

### 2.2 Expression for the Extra Stress Tensor

We employ the Giesekus expression for the stress 31 (Eq. (11.3-1) of Ref. 63):

\[
\tau - \tau_s = \frac{\eta}{4} \frac{d}{dt} \{v \cdot v\} \{R/R\} + \{v \cdot v\} \{R/R\}
\]

\[-\{v \cdot v\} \{R/R\} - \{R/R\} \{v \cdot v\} \]

which when simplified for simple homogeneous shearing flow, we get:

### Table II. Dimensional variables.

<table>
<thead>
<tr>
<th>Variable</th>
<th>Dimensions</th>
<th>Symbol</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time</td>
<td>t</td>
<td>t</td>
</tr>
<tr>
<td>Velocity, ith component</td>
<td>L/t</td>
<td>(u_i)</td>
</tr>
<tr>
<td>Shear rate amplitude</td>
<td>(\dot{y}^0)</td>
<td>(\dot{y}^0)</td>
</tr>
<tr>
<td>Angular frequency</td>
<td>(\omega)</td>
<td>(\omega)</td>
</tr>
<tr>
<td>Relaxation time of fluid</td>
<td>(t)</td>
<td>(t)</td>
</tr>
<tr>
<td>Bead center to center length of rigid dumbbell</td>
<td>L</td>
<td>L</td>
</tr>
<tr>
<td>Bead diameter</td>
<td>L</td>
<td>b</td>
</tr>
<tr>
<td>Cartesian coordinate, distance from stationary plate</td>
<td>L</td>
<td>y</td>
</tr>
<tr>
<td>Cartesian coordinate, flow direction</td>
<td>L</td>
<td>x</td>
</tr>
<tr>
<td>Cartesian coordinate, transverse to flow direction</td>
<td>L</td>
<td>z</td>
</tr>
<tr>
<td>Fluid velocity field</td>
<td>L/t</td>
<td>(v)</td>
</tr>
<tr>
<td>Stress tensor</td>
<td>M/Lt</td>
<td>(\tau)</td>
</tr>
<tr>
<td>Solvent contribution to stress tensor</td>
<td>M/Lt</td>
<td>(\tau_s)</td>
</tr>
<tr>
<td>Orientation vector of a dumbbell with components X,Y,Z</td>
<td>L</td>
<td>(R = r_1 - r_2)</td>
</tr>
<tr>
<td>yz-component of extra stress tensor with components X,Y,Z</td>
<td>M/Lt</td>
<td>(\tau_{yz})</td>
</tr>
<tr>
<td>yz-component of extra stress tensor, solvent</td>
<td>M/Lt</td>
<td>(\tau_{y,s})</td>
</tr>
<tr>
<td>Temperature</td>
<td>T</td>
<td>T</td>
</tr>
<tr>
<td>Bead friction coefficient</td>
<td>M/t</td>
<td>(\zeta = 3\pi \eta)</td>
</tr>
<tr>
<td>Boltzmann constant</td>
<td>M/Lt/V/dumbbell</td>
<td>k</td>
</tr>
<tr>
<td>Steady shear viscosity</td>
<td>M/Lt</td>
<td>(\eta)</td>
</tr>
<tr>
<td>Solvent viscosity</td>
<td>M/Lt</td>
<td>(\eta_s)</td>
</tr>
<tr>
<td>Zero shear rate viscosity</td>
<td>M/Lt</td>
<td>(\eta_0)</td>
</tr>
<tr>
<td>Real part of complex viscosity</td>
<td>M/Lt</td>
<td>(\eta')</td>
</tr>
<tr>
<td>Imaginary part of complex viscosity</td>
<td>M/Lt</td>
<td>(-\eta'')</td>
</tr>
<tr>
<td>mth Fourier loss viscosity</td>
<td>M/Lt</td>
<td>(\eta_{m,loss}(\omega, r^2))</td>
</tr>
<tr>
<td>mth Fourier storage viscosity</td>
<td>M/Lt</td>
<td>(\eta_{m,storage}(\omega, r^2))</td>
</tr>
</tbody>
</table>

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

### Table III. Dimensionless variables and groups.

| Time dependent part of shear rate for velocity profile, assumed linear in \(f(t)\) | \(f(t)\) |
| Weissenberg number, oscillatory shear | \(W_I = \lambda \dot{y}^0\) |
| Deborah number, oscillatory shear | \(D_e = \lambda \omega\) |
| Azimuthal angle | \(\phi\) |
| Polar angle | \(\theta\) |
| Deborah number squared | \(W = (\lambda \omega)^2\) |
| Number of equally-spaced beads on a rod of length \((L-b)\) | \(N\) |
| Number of harmonic | \(n\) |
| Average value | \(\langle \cdot \rangle\) |
| Nabla operator | \(\nabla\) |
| Transpose of a tensor | \(\dagger\) |
| Abbreviation for \(\sin \theta\) in spherical coordinates | \(S\) |
| Abbreviation for \(\sin \phi\) in spherical coordinates | \(s\) |
| Abbreviation for \(\cos \theta\) in spherical coordinates | \(c\) |
| Associated Legendre polynomials | \(P_n^m\) |
| Abbreviation for \(\sin \phi\) in spherical coordinates | \(s_\phi\) |
| Abbreviation for \(\cos \phi\) in spherical coordinates | \(c_\phi\) |
| Orientation distribution | \(\psi(b, \phi, \theta)\) |
| ith term of orientation distribution expansion | \(\psi(b, \phi, \theta)\) |
| Paren functions | \((a, b, \ldots; t)\) |
Then, when replacing the trigonometric functions by the equivalent spherical harmonics (see Eq. (5.5) of Ref. 62 to explore spherical harmonics) we get (Eq. (11.4-2) of Ref. 63):

\[
\frac{\tau_{\psi}}{nkT} = \frac{\lambda}{2} \left[ \frac{d}{dt} \langle \frac{1}{2} \langle P_0^2 \rangle \rangle - \gamma^4 \cos \omega t \langle 2(2P_1^2 - P_2^2) - P_2^2 \rangle \right]
\]

(5)

where the angular brackets denote:

\[
\langle B \rangle = \int_0^{2\pi} \int_0^\pi B(\theta, \phi) \psi(\theta, \phi, t) \sin \theta d\theta d\phi
\]

(6)

where, for a rigid dumbbell suspension in large-amplitude oscillatory shear flow, \(\psi(\theta, \phi, t)\) has been examined previously. Thus, by inserting the expansion for the orientational distribution function (Eq. (26) of Ref. 72 or Eq. (27) of Ref. 75) into Eq. (5), we get:

\[
\frac{\tau_{\psi}}{nkT} = \frac{1}{4\pi} \left[ \frac{d}{dt} \langle \frac{1}{2} \langle P_0^2 \rangle \rangle - \gamma^4 \cos \omega t \\left( \langle 2(2P_1^2 - P_2^2) - P_2^2 \rangle \right) \right]
\]

(7)

Previously, we solved for \(\psi_1, \psi_2\) and \(\psi_3\) (Eq. (34), Eq. (45) and Eq. (54) of Ref. 72), and substituted these into Eq. (7). After rearrangement, we got (Eq. (82) of Ref. 72):

\[
\frac{\tau_{\psi}}{nkT} = \left[ \frac{1}{5(1 + W)} \langle \frac{9}{10(1 + W)(1 + 4W)^2 + 75(1 + 4W)(25 + 9W) + 9(17 - 36W)} \rangle \cos \omega t + \frac{3}{5(1 + W)} \gamma \sin \omega t \right]
\]

(8)

where \(W = (\lambda \omega)^2\). Eq. (8), in the limit as \(\lambda \omega^2\) goes to zero, gives the classic stress response for linear viscoelastic behavior:

\[
\frac{\tau_{\psi}}{nkT} = \left[ \frac{1}{5(1 + W)} \langle \frac{3W}{5(1 + W)} \rangle \cos \omega t + \frac{3}{5(1 + W)} \gamma \sin \omega t \right]
\]

(9)

as it must.

### 3. Method

To get the fifth harmonic, we begin by substituting \(\psi_1, \psi_2\) and \(\psi_3\) (Eq. (34), Eq. (45) and Eq. (54) of Ref. 72) plus our previous expression for \(\psi_4\) (Eq. (163) in Ref. 75) between the square brackets into Eq. (7). We get Eq. (10) of Ref. 32 for the first term. For the second, we get Eq. (11) of Ref. 32, and for the third, Eq. (12) of Ref. 32. Removing the zero contributions in Eqs. (10) through (12) of Ref. 32, we get:

\[
\frac{d}{dt} \left[ \int_0^{2\pi} \int_0^\pi \psi_1^4 \langle \frac{1}{2} \langle P_0^2 \rangle \rangle \sin \theta d\theta d\phi \right] - \frac{1}{10584} \left[ \int_0^{2\pi} \int_0^\pi \psi_1^2 \langle 2(2P_1^2 - P_2^2) - P_2^2 \rangle \sin \theta d\theta d\phi \right]
\]

(10)

and:

\[
\frac{d}{dt} \left[ \int_0^{2\pi} \int_0^\pi \psi_1^4 \langle \frac{1}{2} \langle P_0^2 \rangle \rangle \sin \theta d\theta d\phi \right] - \frac{1}{74088} \left[ \int_0^{2\pi} \int_0^\pi \psi_1^2 \langle 2(2P_1^2 - P_2^2) - P_2^2 \rangle \sin \theta d\theta d\phi \right]
\]

(11)

and:

\[
\frac{d}{dt} \left[ \int_0^{2\pi} \int_0^\pi \psi_1^4 \langle \frac{1}{2} \langle P_0^2 \rangle \rangle \sin \theta d\theta d\phi \right] - \frac{1}{63504} \left[ \int_0^{2\pi} \int_0^\pi \psi_1^2 \langle 2(2P_1^2 - P_2^2) - P_2^2 \rangle \sin \theta d\theta d\phi \right]
\]

(12)

in which \((a, b, ..., t)\) are the paren functions. For instance, the third paren function is given by:
(a, b, c; t) = \frac{1}{\lambda} \int_{-\infty}^{t'} \int_{-\infty}^{t''} e^{-\lambda(t'-\tau)}/\sqrt{t''} e^{-\lambda(t''-\tau)}/\sqrt{t'} \cos \omega t' \cos \omega t'' \cos \omega t''' \mathrm{d}t' \mathrm{d}t'' \mathrm{d}t''' \\
\cos \omega t' \cos \omega t'' \cos \omega t''' \mathrm{d}t' \mathrm{d}t'' \mathrm{d}t''' (13)

Following the method of Bird and Armstrong, we get:

\int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} P_2 \sin \theta \cos \phi \mathrm{d} \theta \mathrm{d} \phi = \frac{48}{5} \pi (14)

\int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} P_0 \cos \phi \cos \phi \cos \phi \cos \phi \sin \theta \cos \phi \mathrm{d} \theta \mathrm{d} \phi = 4\pi (15)

\int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} P_0 \cos \phi \cos \phi \cos \phi \cos \phi \sin \theta \cos \phi \mathrm{d} \theta \mathrm{d} \phi = \frac{4}{5} \pi (16)

\int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} P_2 \cos \phi \cos \phi \cos \phi \cos \phi \sin \theta \cos \phi \mathrm{d} \theta \mathrm{d} \phi = \frac{48}{5} \pi (17)

Substituting these into Eq. (10) through (12) yields Eqs. (21) through (23) of Ref. 32 and then, after rearrangement, we get:

\[ r_{\tau_4} - r_{\tau_4} = \frac{12}{1715} \left[ \lambda \phi \left( \frac{3}{5 \lambda} \frac{d}{dt} (1 - \cos \omega t) \right) - \frac{6}{245} \left( \lambda \phi \right)^3 \left( \frac{23}{3} \left( 0,0,1,1 \right) + 12 \left( \frac{7}{3}, \frac{7}{3}, 1, 1 \right) \right) \right] \cos \omega t \]

(18)

4. RESULTS

4.1 Main Result

Substituting the contributing paren functions (Eqs. (A1) through (A7) of Ref. 72, Eqs. (200) through (217) of Ref. 75) into Eq. (18), and after some factoring, we get, for the first term:

\[ \lambda \phi \left( \frac{3}{5 \lambda} \frac{d}{dt} (1 - \cos \omega t) \right) \]

(19)

For the second term in Eq. (18), we get Eqs. (27) through (29) of Ref. 32 And finally, for the third term, we get Eqs. (29) through (33) of Ref. 32 Substituting Eq. (19) and Eqs. (27) through (33) of Ref. 32 into Eq. (18) and then simplifying gives the polymer contribution to the alternant shear stress response:

where, the zeroth and second order terms of \((\lambda \phi)^n\) match those in Eq. (8), as they should. Eq. (20) is the main result of this paper. Our term of order \((\lambda \phi)^4\), original to this paper, it establishes the first macromolecular connection between the first, third and fifth harmonics of the shear stress.

We can decompose Eq. (20) into its Fourier components as follows:

\[ r_{\tau_4} - r_{\tau_4} = \frac{1}{n \lambda \phi} \left[ \eta_{\phi}^\prime \cos \omega t + \eta_{\phi}^\prime \sin \omega t + \eta_{\phi}^\prime \cos 3\omega t + \eta_{\phi}^\prime \sin 3\omega t + \eta_{\phi}^\prime \cos 5\omega t + \eta_{\phi}^\prime \sin 5\omega t + \cdots \right] \]

(21)

which defines what we call the nth Fourier loss and storage viscosities, \([\eta_{\phi}^\prime (\omega, \gamma), \eta_{\phi}^\prime (\omega, \gamma)\]) for the polymer contribution to the shear stress response of a rigid dumbbell suspension these are given implicitly by comparison of Eq. (21) with Eq. (20). Of course, for the first harmonic, \(\eta_{\phi}^\prime = \eta(\omega, \gamma) - \eta_0\) and \(\eta_{\phi}^\prime = \eta(\omega, \gamma)\). We will use this \([\eta_{\phi}^\prime, \eta_{\phi}^\prime]\) notation to construct Figure 6. The notation of Eq. (21) can be compared or converted to the many other notations for shear stress responses to large-amplitude oscillatory shear flow (see Section 9 of Ref. 51).

4.2 Consistency Checks

4.2.1 Steady shear flow

For steady shear flow, the rigid dumbbell model gives the following expansion for the non-Newtonian viscosity (Eq. (6.7) of Ref. 62, Eq. (11.4-20) of Ref. 63 or Eq. (14.4-18) of Ref. 34):
In the limit, as \( \omega \rightarrow 0 \), Eq. (20) reduces to Eq. (22), as it must.

4.2.2 Intercepts

Evaluating Eq. (20) at \( \omega t = \frac{\pi}{2}, \frac{3\pi}{2} \) gives the ordinate intercepts of the shear stress versus shear rate loops to be used below.

\[
\frac{\tau_{xy} - \tau_{xy,0}}{n k T \lambda_0^4} = \pm \left[ \frac{3\omega}{5(1+W)} \right] \left( \lambda_0^2 \right)^2 \frac{18W(2261 + 3638W + 801W^2)\lambda_0}{175(1+W)^3(1+4W)(1+9W)(25+9W)}
\]

which match those of Eq. (20), as they must. In the limit, as \( \lambda_0^2 \) goes to zero, we get the corresponding loop intercepts for the linear viscoelastic behavior:

\[
\frac{\tau_{xy} - \tau_{xy,0}}{n k T \lambda_0^4} = \pm \left[ \frac{3\omega}{5(1+W)} \right]
\]

which match the intercepts implied by Eq. (9), as they must.

5. DISCUSSION

5.1 Dimensionless Shear Stress Loops

To evaluate Eq. (20), we begin by following Dealy et al. (1973) in plotting loops of shear stress versus shear rate, since these best bring out distortions from ellipticity.\(^{35-37}\) Figure 3 through Figure 5 show the improvements to Eq. (8) afforded by the new \( (\lambda_0^2)^2 \) term from our expansion of the orientational distribution function (Eq. (163) of Ref. 75). Figure 3 through Figure 5 thus also show the improvements to Eq. (8) afforded by the new \( (\lambda_0^2)^2 \) term in Eq. (20). Whereas at low Deborah number, we see no improvement (Figure 3), at a Deborah number of one, we see significant improvement (Figure 4). At much higher Deborah number, we see significant improvement, unless the Weissenberg number is also high (Figure 5). Our \( (\lambda_0^2)^2 \) term expansions thus break down when both the Weissenberg and the Deborah numbers are high.

Following Philippoff’s Figure 4 of Ref. 38, we construct Figure 6, the Fourier decomposition of the polymer contribution to the alternant shear stress response. From Figure 6 we learn that the signed area of the loops for harmonics higher than the first is zero, as it should be, and thus, that all of the polymer contribution to the viscous dissipation comes from \( \eta_p \) (see after Eq. (17) of Ref. 39 and after Eq. (9) of Ref. 40; also, compare columns 3 and 4 of Table II of Ref. 41).

5.2 Comparison with Continuum Model

To deepen our understanding of our main result, we next follow Bird et al.\(^{72}\) and compare our main result, Eq. (20) with the corotational Maxwell model, the simplest relevant two-constant (\( \eta_0 \) and \( \lambda_0 \)) continuum model (see last paragraph of INTRODUCTION). By relevant we mean that the model predicts higher harmonics. Unlike the rigid dumbbell model, the corotational Maxwell model ignores the association between the molecular structural changes and the rheological behavior. Instead the corotational Maxwell model is a sophisticated guess based on some knowledge of the measured behavior of the polymeric liquids.\(^{42}\)

We write the shear stress response for the corotational Maxwell model, up to fourth order of \( (\lambda_0^2)^2 \) contributions, as \([\text{Eq. (58) of Ref. 51}]:\)

\[
\eta_p(\dot{\gamma}) - \eta_0 = -\frac{\tau_{xy} - \tau_{xy,0}}{\dot{\gamma}} = \frac{n k T \lambda_0^4}{1 + W} \left[ \frac{3\omega}{5(1+W)} \right] \left( \lambda_0^2 \right)^2 \frac{18W(2261 + 3638W + 801W^2)\lambda_0}{175(1+W)^3(1+4W)(1+9W)(25+9W)}
\]

which thus includes odd harmonics up to and including the fifth.

If we identify \( n k T \lambda_0^4 \) on the left side of Eq. (20) with \( \eta_0 \), by considering the special case where \( \eta_p \ll \eta_0 \) in Eq. (20) so that \( \eta_p = \eta_0 \), we can then compare with the corotational Maxwell
solution, Eq. (25).

For small-amplitude oscillatory shear, whereas for rigid dumbbells we get (Eqs. (84) and (85) of Ref. 72):

\[ \frac{\eta'(\lambda \omega)}{nkT \lambda} = 1 - \frac{3}{5} \left[ \frac{W}{1+W} \right] \]  
(26)

and:

\[ \frac{\eta''(\lambda \omega)}{nkT \lambda} = \frac{3}{5} \left[ \frac{1}{1+W} \right] \lambda \omega \]  
(27)

for the corotational Maxwell model we get (Eqs. (86) and (87) of Ref. 72):

\[ \frac{\eta'(\lambda \omega)}{nkT \lambda} = \frac{1}{1+W} \]  
(28)

and:

\[ \frac{\eta''(\lambda \omega)}{nkT \lambda} = \frac{1}{1+W} \lambda \omega \]  
(29)

Fig. 3. Counterclockwise loops of minus dimensionless shear stress versus dimensionless shear rate calculated for the rigid dumbbell model: [Eq. (20), solid curves] versus [Eq. (8), dotted curves] by increasing the Weissenberg number with a fixed Deborah number of 0.1.

Fig. 4. Counterclockwise loops of minus dimensionless shear stress versus dimensionless shear rate calculated for the rigid dumbbell model: [Eq. (20), solid curves] versus [Eq. (8), dotted curves] by increasing the Weissenberg number with a fixed Deborah number of 10.

Fig. 5. Counterclockwise loops of minus dimensionless shear stress versus dimensionless shear rate calculated for the rigid dumbbell model: [Eq. (20), solid curves] versus [Eq. (8), dotted curves] by increasing the Weissenberg number with a fixed Deborah number of 10.

Fig. 6. Fourier decomposition of the polymer contribution to the alternant shear stress response of rigid dumbbell suspension [Eq. (20)] into the first \( m = 1 \), third \( m = 3 \) and fifth \( m = 5 \) harmonics. Axis label following notation of Eq. (21).
For linear viscoelasticity of both the rigid dumbbell and the corotational Maxwell models, \( \eta'(\omega) \) and \( \eta''(\omega) \) predictions are qualitatively alike (see Figure 6 of Ref. 72).

### 5.2.1 First harmonic

We next examine the nonlinear behavior of the first harmonic. Whereas for the rigid dumbbell model, from Eq. (20), we get:

\[
\eta'(\omega, \omega') = \frac{1}{\eta} \left[ \frac{2}{1 + W} \right] \left( \lambda \omega \right)^{\frac{5}{4}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{15}{8}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{1}{2}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{1}{4}} \lambda \omega
\]

and:

\[
\eta''(\omega, \omega') = \frac{1}{\eta} \left[ \frac{2}{1 + W} \right] \left( \lambda \omega \right)^{\frac{5}{4}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{15}{8}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{1}{2}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{1}{4}} \lambda \omega
\]

Eqs. (30) and (31) correspond to Eqs. (88) and (89) of Ref. 72 plus the new \((\lambda \omega)^4\) term from Eq. (20). Figure 7 and Figure 8 show the improvements afforded by the new \((\lambda \omega)^4\) term in Eq. (20), and thus, the corresponding improvements to Eqs. (9.3) and (9.4) of Ref. 62. Figure 7 and Figure 8 thus also show the improvements afforded by the new \((\lambda \omega)^4\) term from our expansion of the orientational distribution function (Eq. (163) of Ref. 75), and thus, the corresponding improvement to Eq. (9.2) of Ref. 62.

For the first harmonic, for the rigid dumbbell model, the coefficients of \((\lambda \omega)^4\) in the expressions for \(\eta'(\omega, \omega')\) or for \(\eta''(\omega, \omega')\) are given by Eqs. (88) and (89) of Ref. 72. For the corotational Maxwell model, the corresponding coefficients are given by Eqs. (90) and (91) of Ref. 72. Figure 7 of Ref. 72 shows that for these coefficients of \((\lambda \omega)^4\) the predictions are qualitatively alike.

Figure 9 compares the rigid dumbbell model versus the corotational Maxwell model first harmonic coefficients of \((\lambda \omega)^4\) in the expressions for \(\eta'(\omega, \omega')\) [Eq. (30)] versus Eq. (32), solid curves] and for \(\eta''(\omega, \omega')\) [Eq. (31)] versus Eq. (33), dashed curves. We find that the predicted behaviors of these coefficients of \((\lambda \omega)^4\) for the molecular and continuum models are qualitatively alike.

### 5.2.2 Third harmonic

We next examine the behavior of the third harmonic. Whereas for the rigid dumbbell model, from Eq. (20), we get for the coefficient of \((\lambda \omega)^4\) in the expressions for the parts of the shear stress response that are in-phase with cos \(3\omega t\):

\[
\eta'(\omega, \omega') = \frac{1}{\eta} \left[ \frac{2}{1 + W} \right] \left( \lambda \omega \right)^{\frac{5}{4}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{15}{8}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{1}{2}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{1}{4}} \lambda \omega
\]

and with \(\lambda \omega\):

\[
\left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{5}{4}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{15}{8}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{1}{2}} \left[ \frac{1}{1 + W} \right] \left( \lambda \omega \right)^{\frac{1}{4}} \lambda \omega
\]

Eqs. (34) and (35) correspond to Eqs. (88) and (89) of Ref. 72 plus the new \((\lambda \omega)^4\) term from Eq. (20). Figure 7 and Figure 8 show the improvements afforded by the new \((\lambda \omega)^4\) term in Eq. (20), and thus, the corresponding improvements to Eqs. (9.3) and (9.4) of Ref. 62. Figure 7 and Figure 8 thus also show the improvements afforded by the new \((\lambda \omega)^4\) term from our expansion of the orientational distribution function (Eq. (163) of Ref. 75), and thus, the corresponding improvement to Eq. (9.2) of Ref. 62.

For the third harmonic, for the rigid dumbbell model, the coefficients of \((\lambda \omega)^4\) in the expressions for the parts of the shear stress response that are in-phase with \(3\omega t\) or with \(\sin 3\omega t\) are given by Eqs. (92) and (93) of Ref. 72. For the corotational Maxwell model, the corresponding coefficients are given by Eqs. (94) and (95) of Ref. 72. Figure 8 of Ref. 72 shows that for these coefficients of \((\lambda \omega)^4\) the predictions are qualitatively alike.

Figure 10 compares the rigid dumbbell model versus the corotational Maxwell model third harmonic coefficients...
of \((\lambda \gamma^0)^4\) in the expressions for the parts of the shear stress response that are in-phase with \(\cos 3\omega t\) [Eq. (34) versus Eq. (36), solid curves] and with \(\sin 3\omega t\) [Eq. (35) versus Eq. (37), dashed curves]. As for the first harmonic (see Figure 9), we find that the predicted behaviors of these coefficients of \((\lambda \gamma^0)^4\) for the molecular and continuum models are qualitatively alike.

### 5.2.3. Fifth harmonic

We next examine the behavior of the fifth harmonic. Whereas for the rigid dumbbell model, from Eq. (20), we get for the coefficient of \((\lambda \gamma^0)^4\) in the expressions for the parts of the shear stress response that are in-phase with \(\cos 5\omega t\):

\[
\left\{ \begin{array}{c}
5525 - 149191W - 290928W^2 \\
-118044W^3 + 87480W^4
\end{array} \right\}
\]

\[
\frac{1}{154(1+W)(1+4W)(1+9W)} \frac{1}{(25+9W)(1+16W)(100+81W)}
\]

(38)

and with \(5\omega t\):

\[
\lambda \omega \left\{ \begin{array}{c}
-516425 + 725992W \\
+3260205W^2 + 2710908W^3
\end{array} \right\}
\]

\[
\frac{-3}{154(1+W)(1+4W)(1+9W)} \frac{-3}{(25+9W)(1+16W)(100+81W)}
\]

(39)

for the corotational Maxwell model, from Eq. (58) of Ref. 51 (or from Eq. (25) above) we get for the coefficient of \((\lambda \gamma^0)^4\) in

![Fig. 7. Behavior of \(\eta(\lambda \gamma^0, \lambda \gamma^0)\) at different \(\lambda \gamma^0\) values [Eq. (30), solid curves] versus the first two terms of Eq. (30) [Eq. (88) of 72, dashed curves] for the rigid dumbbell model.](image)

![Fig. 8. Behavior of \(\eta(\lambda \gamma^0, \lambda \gamma^0)\lambda \omega\) at different \(\lambda \gamma^0\) values [Eq. (31), solid curves] versus first two terms of Eq. (31) [Eq. (89) of 72, dashed curves] for the rigid dumbbell model.](image)

![Fig. 9. The rigid dumbbell model (blue) versus the corotational Maxwell model (red) for large-amplitude oscillatory shear flow: Coefficients of \((\lambda \gamma^0)^4\) in expressions for \(\eta(\omega \gamma^0, \gamma^0)\) [Eq. (30) versus Eq. (32), solid curves] and for \(\eta(\omega \gamma^0, \gamma^0)\) [Eq. (31) versus Eq. (33), dashed curves].](image)

![Fig. 10. The rigid dumbbell model (blue) versus the corotational Maxwell model (red) for large-amplitude oscillatory shear flow: Coefficients of \((\lambda \gamma^0)^4\) in expressions for \(\cos 3\omega t\) [Eq. (34) versus Eq. (36), solid curves] and for \(\sin 3\omega t\) [Eq. (35) versus Eq. (37), dashed curves].](image)
the expressions for the parts of the shear stress response that are in-phase with \( \cos 5\omega t \):

\[
\frac{1 - 85W + 274W^2}{16(1 + W)^3(1 + 4W)(1 + 9W)(1 + 16W)(1 + 25W)} \quad (40)
\]

and with \( \sin 5\omega t \):

\[
\frac{1 - 15W + 8W^2}{16(1 + W)^3(1 + 4W)(1 + 9W)(1 + 16W)(1 + 25W)} \lambda \omega \quad (41)
\]

Figure 11 gives us our first glimpse of the behavior of a fifth harmonic derived from macromolecular theory. Specifically, Figure 11 compares the rigid dumbbell model versus the corotational Maxwell model fifth harmonic coefficients of \((\lambda \omega)^4\) in the expressions for the parts of the shear stress response that are in-phase with \( \cos 5\omega t \) [Eq. (38) versus Eq. (40), solid curves] and with \( \sin 5\omega t \) [Eq. (39) versus Eq. (41), dashed curves]. As for the first and third harmonics (see Figure 9 and Figure 10), we find that the predicted behaviors of these coefficients of \((\lambda \omega)^4\) for the molecular and continuum models are qualitatively alike.

6. CONCLUSION

In this work, we derived the polymer contribution to the shear stress response up to and including its fifth harmonic [Eq. (20)] for a suspension of rigid dumbbells. For this, we used an extended expansion of the orientation distribution, to include the fourth order term (Eq. (163) of Ref. 74). We then used this extension to calculate the polymer contribution to the shear stress response. We thus succeeded in employing the general method of Bird and Armstrong \(^{31}\) to extend our previous work.\(^{12,74}\) Our expression for the fifth harmonic, Eq. (20), is the only one to have been derived from a molecular theory. Our paper thus provides the first glimpse of the molecular origins of a shear stress harmonic higher than the third. We find the behavior of this fifth harmonic predicted from molecular (rigid dumbbell) and from continuum (corotational Maxwell) theories to be qualitatively alike (see Figure 11).

Our analysis has also provided next-term improvements to previous approximate expressions for the first and third harmonics, for the rigid dumbbell suspension. From the first harmonics, we get Eqs. (30) and (31) for the material functions \( \eta'(\lambda \omega, \lambda \omega) \) and \( \eta''(\lambda \omega, \lambda \omega) \). These are significant improvements to the classic expressions of Bird, Warner and Evans (Eqs. (88) and (89) of Ref. 72) for \( \eta'(\lambda \omega, \lambda \omega) \) and \( \eta''(\lambda \omega, \lambda \omega) \) for rigid dumbbell theory.

The rigid dumbbell model gives qualitatively correct results for many rheological functions, for both solutions and polymer melts, indicating that it is the orientation of the constituent molecules that is more important than the detailed motion of the individual parts of the polymer chains.\(^{42}\) In other words, experimental verification of the coefficients in Eq. (20) can be approached with either dilute or concentrated polymer solutions, or even with melts.

We close with the suggestion that, with the help of Eqs. (6) and (7) of Ref. 72, our main result [Eq. (20)] can be used to inform the design (or re-design) of macromolecules to be incorporated in elastic liquids for viscous dampers\(^{43-46}\) (Section 3.3 of Ref. 47; Section 8.7 of Ref. 48; Problem 5.5 of Ref. 49-50).

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ost + $\cos 2\omega t$}; Eq. (6.41b) should be $p_{11} = \alpha (\cos \omega t + \sin \omega t)$; in line 4 of p. 113, $\alpha \cos \omega t$ should be $\alpha \cos \omega t$; in the sentence preceding Eq. (6.43), and also in Eq. (6.43), “the out-of-phase part of $p_{11}$” should be “the part of $p_{11}$ that is in-phase with $\phi$”.  
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