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Inertial effects on the rheology of a dilute emulsion

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The behaviour of an isolated nearly spherical drop in an ambient linear flow is examined analytically at small but finite Reynolds numbers, and thereby the first effects of inertia on the bulk stress in a dilute emulsion of neutrally buoyant drops are calculated. The Reynolds numbers, $Re=\dot{\gamma}a^2\rho/\mu$ and $\hat{Re}=\dot{\gamma}a^2\rho/\hat{\mu}$, are the relevant dimensionless measures of inertia in the continuous and disperse (drop) phases, respectively. Here, a is the drop radius, $\dot{\gamma}$ is the shear rate, ρ is the common density and $\hat{\mu}$ and μ are, respectively, the viscosities of the drop and the suspending fluid. The assumption of nearly spherical drops implies the dominance of surface tension, and the analysis therefore corresponds to the limit of the capillary number (Ca) based on the viscosity of the suspending fluid being small but finite; in other words, $Ca \ll 1$, where $Ca=\mu a\dot{\gamma}/T$, T being the coefficient of interfacial tension. The bulk stress is determined to $O(\phi Re)$ via two approaches. The first one is the familiar direct approach based on determining the force density associated with the disturbance velocity field on the surface of the drop; the latter is determined to $O(Re)$ from a regular perturbation analysis. The second approach is based on a novel reciprocal theorem formulation and allows the calculation, to $O(Re)$, of the drop stresslet, and hence the emulsion bulk stress, with knowledge of only the leading-order Stokes fields. The first approach is used to determine the bulk stress for linear flows without vortex stretching, while the reciprocal theorem approach allows one to generalize this result to any linear flow. For the case of simple shear flow, the inertial contributions to the bulk stress lead to normal stress differences (N_1, N_2) at $O(\phi Re)$, where ϕ ($\ll 1$) is the volume fraction of the disperse phase. Inertia leads to negative and positive contributions, respectively, to N_1 and N_2 at $O(\phi Re)$. The signs of the inertial contributions to the normal stress differences may be related to the $O(ReCa)$ tilting of the drop towards the velocity gradient direction. These signs are, however, opposite to that of the normal stress differences in the creeping flow limit. The latter are $O(\phi Ca)$ and result from an $O(Ca^2)$ deformation of the drop acting to tilt it towards the flow axis. As a result, even a modest amount of inertia has a significant effect on the rheology of a dilute emulsion. In particular, both normal stress differences reverse sign at critical Reynolds numbers (Re_c) of $O(Ca)$ in the limit $Ca \ll 1$. This criterion for the reversal in the signs of N_1 and N_2 is more conveniently expressed in terms of a critical Ohnesorge number (Oh) based on the viscosity of the suspending fluid, where $Oh=\mu/(\rho a T)^{1/2}$. The critical Ohnesorge number for a sign reversal in N_1 is found to be lower than that for N_2, and the precise numerical value is a function of λ. In uniaxial extensional flow,

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the Trouton viscosity remains unaltered at $O(\phi Re)$, the first effects of inertia now being restricted to $O(\phi Re^{3/2})$. The analytical results for simple shear flow compare favourably with the recent numerical simulations of Li & Sarkar (*J. Rheol.*, vol. 49, 2005, p. 1377).

1. Introduction

Emulsions play a critical role in numerous industrial processes, and in many of these applications, a knowledge of their rheological properties is an obvious prerequisite to predicting their flow behaviour. In this regard, we derive the constitutive equation, accurate to $O(\phi Re)$, for a dilute emulsion of neutrally buoyant nearly spherical drops subject to an ambient linear flow. Here, ϕ is the volume fraction of the drop phase, and $Re = \dot{\gamma} a^2 \rho / \mu$ is the relevant dimensionless measure of inertia for flow around a single drop of radius a, where $\dot{\gamma}$ is an appropriate shear rate, ρ is the density and μ and $\hat{\mu}$ are the continuous and disperse phase viscosities; both phases are assumed to exhibit a Newtonian rheology. One may also define a Reynolds number, $\hat{Re} = \dot{\gamma} a^2 \rho / \hat{\mu}$, for the flow inside a drop with $\hat{Re} = Re/\lambda$, $\lambda = \hat{\mu}/\mu$ being the viscosity ratio. The analysis in this paper is restricted to the limit where both ϕ and Re (and $\hat{Re}$) are small but finite, and in addition, interfacial tension forces are assumed to be strong enough to keep the drops nearly spherical. The latter assumption implies that the capillary number, $Ca = \mu a \dot{\gamma} / T$, is small compared to unity, where T is the coefficient of interfacial tension. Thus, the $O(\phi Re)$ correction to the constitutive relation for a dilute emulsion is related to the effect of weak inertia on the flow field around a single spherical drop in a region of the order of its own size; the analysis here neglects hydrodynamic interactions between two or more drops that might become important at higher volume fractions.

There have been earlier attempts to characterize the role of inertia in the rheology of a suspension of rigid particles: to $O(\phi Re)$ for a general linear flow and to $O(\phi Re^{3/2})$ for simple shear flow. For both suspensions and emulsions, the theoretical calculation of the inertial correction at $O(\phi Re^{3/2})$ is a more difficult task than the one at $O(\phi Re)$, the difficulty being related to the non-uniformity of the leading-order Stokes approximation. Unlike the $O(\phi Re)$ correction, the dominant contributions to the bulk stresses at $O(\phi Re^{3/2})$ originate from regions relatively remote from the particle or drop on the microscale. Lin, Peery & Schowalter (1970) were the first to investigate the rheology of an inertial suspension in the infinitely dilute limit; they obtained results accurate to $O(\phi Re^{3/2})$ for simple shear flow via a traditionally matched asymptotic expansions approach. Later, Stone, Brady & Lovalenti (2000) redid the calculation via an alternate and more concise formulation in Fourier space based on the generalized reciprocal theorem, and in the process, also extended the rheological results at $O(\phi Re)$ to an arbitrary ambient linear flow. In a forthcoming publication (Subramanian & Koch 2010), it has been shown that these earlier calculations for the suspension bulk stress are incomplete since they neglect the contributions of the Reynolds stresses at $O(\phi Re^{3/2})$. However, the predictions at $O(\phi Re)$ are correct and have also since been verified numerically; the numerical calculations extend the theoretical predictions to finite Re (see Mikulencak & Morris 2004). Inertia leads to a non-Newtonian suspension rheology at $O(\phi Re)$, and for simple shear flow in particular, there arise normal stress differences at this order although the shear viscosity remains unaltered. However, this paper not only focuses on the qualitative change in rheology in the mere presence of microscale inertia (which is the case for both suspensions and emulsions), but also changes in rheology that depend crucially on the strength of inertial effects relative to viscous forces. The latter situation is specific to an emulsion. A dilute emulsion exhibits a non-Newtonian rheology even in the inertialess limit. For finite Re, both inertial and viscous forces compete with the restoring interfacial tension forces that arise because of drop deformation.

Inertial effects become significant in emulsions with relatively low viscosity suspending fluids at high shear rates, turbulent flows being an example. Consider a turbulent flow of an aqueous emulsion ($\rho \approx 10^3$ kg m^{-3}, $\mu \approx 10^{-3}$ Pa s^{-1}) with a drop size of about 50 µm. With a typical estimate for the dissipation rate per unit volume, $\epsilon \approx 10$ cm^2 s^{-3}, one obtains the Kolmogorov length scale as $l_\kappa = (\epsilon^3/\nu)^{1/4} \approx 170$ µm and the corresponding shear rate, $\dot{\gamma}_\kappa = (\epsilon/\nu)^{1/2} \approx 25$ s^{-1}. The drops being much smaller than the Kolmogorov scale, the ambient flow seen by a single drop is a nearly linear flow with a fluctuating velocity gradient tensor. The relevant dimensionless measure of the drop scale inertia is therefore the Reynolds number based on the drop size and the Kolmogorov shear rate, being given by $Re = a^2\epsilon^{1/2}/\nu^{3/2} \approx 0.08$ (the turbulent flow, even on the scale of a micron-sized drop, is not exactly linear; a deviation of the ambient flow from linearity, at $O(Re)$, is accounted for in the present analysis). The smallness of Re, that is, the fact that $a^2/\nu \ll (\nu/\epsilon)^{1/2}$ also shows turbulence to be a slowly evolving flow on the microscale. For the same parameter values, one finds $Ca = a(\epsilon\mu\rho)^{1/2}/T \approx 10^{-4}$ with $T \approx 10$ mN m^{-1}, implying that the drops will remain approximately spherical as is assumed in the following analysis. Thus, in this parameter regime, one expects the microscale inertia induced alteration of emulsion rheology, and the resulting flow behaviour, to be much more important than the well-known non-Newtonian response related to drop deformation induced by purely viscous forces ($Re = 0$). The present effort will help further the understanding of the flow behaviour of emulsions in turbulent flow, or more generally, the behaviour of disperse multiphase systems in flows that remain rheologically complex (non-viscometric) down to the scale of the disperse phase constituents.

The principal result of this paper is an expression for the bulk stress in a dilute emulsion, to $O(\phi Re)$, in a general linear flow of the form $\boldsymbol{u}^\infty = \boldsymbol{\Gamma}\cdot\boldsymbol{x} = (\boldsymbol{E} + \boldsymbol{\Omega})\cdot\boldsymbol{x}$, where $\boldsymbol{\Gamma}$ is the transpose of the velocity gradient tensor, and $\boldsymbol{E}$ and $\boldsymbol{\Omega}$ are the ambient rate of strain tensor and the transpose of the vorticity tensor, respectively. One finds

$$\begin{aligned}\Sigma_{ij} = -p_t\,\delta_{ij} + \mu\Bigg\{ 2E_{ij} + 2\phi\left[\frac{(5\lambda+2)}{2(\lambda+1)}E_{ij} + \frac{1}{10}\nabla^2 E_{ij}\right]_{\boldsymbol{x}_d(t)} \\ + (\phi Re)\left[\frac{(27\lambda^2+30\lambda+10)}{15(\lambda+1)^2}\frac{\mathrm{D}^\infty E_{ij}}{\mathrm{D}t} - \frac{4(3\lambda^2+3\lambda+1)}{9(\lambda+1)^2}(\Omega_{ik}E_{jk} + \Omega_{jk}E_{ik})\right]_{\boldsymbol{x}_d(t)} \\ + O(\phi^2, \phi Re^{3/2}, \phi Ca)\Bigg\}, \end{aligned} \qquad (1.1)$$

where p_t is an arbitrary isotropic pressure, $\boldsymbol{x}_d(t)$ denotes the position of the translating drop relative to a laboratory reference frame, and $\mathrm{D}^\infty/\mathrm{D}t = \partial/\partial t + \boldsymbol{u}^\infty\cdot\nabla$ denotes the material derivative associated with the ambient flow. As for the case of rigid particle suspensions above, inertia evidently leads to a non-Newtonian emulsion rheology. The arbitrariness referred to above is that of the total pressure in the emulsion which is, of course, determined by incompressibility. However, a disperse phase pressure may still be extracted from a constitutive relation. The normal stresses induced by the disperse phase are thought to play a crucial role in the shear-induced migration associated with irreversible interparticle interactions (see Nott & Brady 1994; Morris & Brady 1998). The present effort, however, focuses on the deviatoric part of the stress contributed by isolated drops in a dilute emulsion and the resulting implications for rheology.

For simple shear ($\boldsymbol{E} = (1/2)(\mathbf{1}_x\mathbf{1}_y + \mathbf{1}_y\mathbf{1}_x)$, $\boldsymbol{\Omega} = (1/2)(\mathbf{1}_x\mathbf{1}_y - \mathbf{1}_y\mathbf{1}_x)$ with x, y and z corresponding to the flow, gradient and vorticity directions, respectively), one finds that the viscosity is unchanged to $O(\phi Re)$, but that the inertial contributions to the normal stress differences at this order have signs opposite to the corresponding normal stress differences resulting from drop deformation in the absence of inertia (e.g. see Schowalter, Chaffey & Brenner 1968). The latter are $O(\phi Ca)$ and, therefore, even a modest amount of inertia ($Re \sim Ca$) qualitatively changes the rheology of a dilute emulsion by reversing the sign of one or both normal stress differences. Using (1.1), and including the $O(\phi Ca)$ contributions, the expressions for N_1 and N_2, non-dimensionalized by $\mu\dot{\gamma}$, are given by

$$N_1 = \phi Ca\, \frac{1}{40}\left(\frac{19\lambda + 16}{\lambda+1}\right)^2 - \phi Re \frac{4(3\lambda^2 + 3\lambda + 1)}{9(\lambda + 1)^2} + O(\phi^2, \phi Ca^2, \phi Re^{3/2}), \quad (1.2)$$

$$N_2 = -\phi Ca\, \frac{(551\lambda^3 + 1623\lambda^2 + 1926\lambda + 800)}{280(\lambda + 1)^3} + \phi Re \frac{2(3\lambda^2 + 3\lambda + 1)}{9(\lambda + 1)^2} + O(\phi^2, \phi Ca^2, \phi Re^{3/2}), \quad (1.3)$$

where the $O(\phi Re)$ contributions in both N_1 and N_2, in the limit $\lambda \to \infty$, reduce to those known for a rigid particle suspension at this order (Lin *et al.* 1970). The critical condition for a reversal in sign of either N_1 or N_2 is related to the ratio (Re/Ca), and is thus independent of the imposed shear rate. It is therefore best characterized in terms of a dimensionless number that is a function of the system properties, but not the flow. The Ohnesorge number (Oh), defined as $Oh = (Ca/Re)^{1/2} = \mu/(\rho a T)^{1/2}$, is such a parameter, and one obtains the following critical Ohnesorge numbers for N_1 and N_2:

$$Oh_c^{N1} = \frac{4\sqrt{10}}{3}\frac{(3\lambda^2 + 3\lambda + 1)^{1/2}}{(19\lambda + 16)}, \quad (1.4)$$

$$Oh_c^{N2} = \frac{4}{3}\left[\frac{35(\lambda + 1)(3\lambda^2 + 3\lambda + 1)}{(551\lambda^3 + 1623\lambda^2 + 1926\lambda + 800)}\right]^{1/2}. \quad (1.5)$$

For steady extensional flow ($\boldsymbol{\Gamma} = \boldsymbol{E}$, $\boldsymbol{\Omega} = \mathbf{0}$), it is readily seen from (1.1) that the inertial correction is identically zero. Thus, rather remarkably, inertia leaves the extensional viscosity unaltered, to $O(\phi Re)$, irrespective of the viscosity ratio. The first effects of inertia in this case enter at $O(\phi Re^{3/2})$ and, as indicated earlier, a calculation of the same requires a singular perturbation analysis. The present analysis was motivated in part by the results of earlier simulations carried out by Li & Sarkar (2005), which indicated the existence of such a reversal in the sign of the normal stress differences. The authors simulated a dilute emulsion of unit viscosity ratio by placing a spherical drop between parallel plates with periodic boundary conditions imposed in the horizontal directions. The analytical predictions will be compared with the numerical results in § 5.

The threshold conditions (1.4) and (1.5) are better understood in the context of a specific system. Considering a typical low viscosity organic fluid with $\rho \approx 10^3\,\mathrm{kg\,m^{-3}}$, $\mu \approx 5 \times 10^{-3}\,\mathrm{Pa\,s^{-1}}$, and a coefficient of interfacial tension, $T \approx 10^{-2}\,\mathrm{N m^{-1}}$, again typical for organic systems, one obtains the Ohnesorge number as a function of the drop size: $Oh \approx 1.58 \times 10^{-3}/a^{1/2}$. Now, for an emulsion with similar disperse and continuous phase viscosities ($\lambda \approx 1$), one has $Oh_c^{N1} \approx 0.32$ and $Oh_c^{N2} \approx 0.42$. This system must therefore exhibit a positive N_1 and a negative N_2 for drops smaller than 14 µm, a positive N_1 and a positive N_2 for drop sizes between 14 µm and 24 µm, and finally, a negative N_1 and a positive N_2 for drop sizes greater than 24 µm when inertial stresses become dominant.

The paper is organized as follows. In § 2 we consider a density-matched spherical Newtonian drop in an ambient linear flow of a Newtonian suspending fluid and determine the velocity and pressure fields in either phase, accounting for the first effects of inertia, via a regular perturbation approach. Later, in § 3, the corresponding stress field in the suspending fluid, at $O(Re)$, is used to determine the bulk stress, to $O(\phi Re)$, in a dilute emulsion subject to a linear flow without vortex stretching. The calculations in this direct approach to the stress determination are tedious; the most involved part of the entire calculation by far is the $O(Re)$ correction to the stresslet. Thus, in § 4, we again calculate the stresslet to $O(Re)$ via an alternate approach based on a novel reciprocal theorem formulation. This approach is shown to yield the same result, thereby validating the result for the $O(Re)$ disturbance velocity fields derived via the straightforward yet cumbersome perturbation analysis of § 2. Furthermore, the reciprocal theorem approach allows one to generalize the result for the bulk stress, to $O(\phi Re)$, to an arbitrary linear flow. The rheology of a dilute emulsion in the inertialess limit, to $O(\phi Ca)$, is well known (e.g. see Frenkel & Acrivos 1970; Schowalter *et al.* 1968). For the case of simple shear flow, an inertialess emulsion exhibits a positive first normal stress difference (N_1) and a negative second normal stress difference (N_2), both at $O(\phi Ca)$, on account of drop deformation. Combining our calculations for the inertial contributions with the known expression for the bulk stress in the creeping flow limit, we show in § 5 that inertia qualitatively alters the emulsion rheology in simple shear when $Re \sim O(Ca)$. This is due to changes in sign of both N_1 and N_2, and as explained above, the threshold conditions for the reversals in sign are best formulated in terms of a critical Ohnesorge number for each of N_1 and N_2. On the other hand, inertial effects leave the extensional rheology of a dilute emulsion unaltered to $O(\phi Re)$. It is argued thereafter that the results here are expected to hold even for surfactant-laden interfaces, at least in the limit of highly elastic interfaces. The theoretical predictions are finally compared with recent simulation results. As is implicit in the above scalings, in all calculations we assume the emulsion to still be dilute enough for hydrodynamic interactions between drops to be unimportant. Pair interactions lead to an $O(\phi^2)$ contribution to the bulk stress in the limit of small ϕ, and the latter remains smaller than the contributions of $O(\phi Re)$ and $O(\phi Ca)$ included in this paper provided $\phi \ll Re, Ca$. Finally, in § 6 we present a brief summary of our results.

2. The velocity and pressure fields due to a neutrally buoyant drop in a simple shear flow

In this section, we solve the velocity and pressure fields owing to a neutrally buoyant Newtonian drop of viscosity $\hat{\mu}$ suspended in a simple shearing flow of a Newtonian suspending fluid with viscosity μ in the limit of small but finite inertia. Using the familiar scalings, viz. the drop radius a for position ($\boldsymbol{r}$), the reciprocal of the ambient shear rate $\dot{\gamma}^{-1}$ for time, $\dot{\gamma}a$ for the velocities, $\hat{\mu}\dot{\gamma}$ for the stresses within the drop and $\mu\dot{\gamma}$ for stresses in the exterior fluid, one obtains the non-dimensionalized governing equations in the exterior and interior of the drop at steady state. In a coordinate system with its origin at the centre of the drop, the equations

$$\nabla^2 \boldsymbol{u} - \nabla p = Re\, \boldsymbol{u}\cdot\nabla\boldsymbol{u}, \tag{2.1}$$

$$\nabla\cdot\boldsymbol{u} = 0 \tag{2.2}$$

govern the fluid motion outside the drop, and

$$\nabla^2 \hat{\boldsymbol{u}} - \nabla \hat{p} = \hat{Re}\, \hat{\boldsymbol{u}}\cdot\nabla\hat{\boldsymbol{u}}, \tag{2.3}$$

$$\nabla\cdot\hat{\boldsymbol{u}} = 0, \tag{2.4}$$

govern the fluid motion within the drop. Here, $(\hat{\boldsymbol{u}}, \hat{p})$ and $(\boldsymbol{u}, p)$, respectively, denote the interior and exterior velocity and pressure fields, while $\hat{Re} = \dot{\gamma}a^2\rho/\hat{\mu}$ and $Re = \dot{\gamma}a^2\rho/\mu$ are corresponding dimensionless measures of the importance of inertia. We impose the following boundary conditions at the surface of the undeformed drop $(r = 1)$:

$$\boldsymbol{u} = \hat{\boldsymbol{u}}, \tag{2.5}$$

$$\boldsymbol{u}\cdot\boldsymbol{n} = 0, \tag{2.6}$$

$$(\boldsymbol{\sigma}\cdot\boldsymbol{n}) - (\boldsymbol{\sigma}:\boldsymbol{nn})\boldsymbol{n} = \lambda[(\hat{\boldsymbol{\sigma}}\cdot\boldsymbol{n}) - (\hat{\boldsymbol{\sigma}}:\boldsymbol{nn})\boldsymbol{n}], \tag{2.7}$$

where $\boldsymbol{\sigma} = -p\,\boldsymbol{I} + (\nabla\boldsymbol{u} + \nabla\boldsymbol{u}^{\dagger})$ and $\hat{\boldsymbol{\sigma}} = -\hat{p}\,\boldsymbol{I} + (\nabla\hat{\boldsymbol{u}} + \nabla\hat{\boldsymbol{u}}^{\dagger})$ are, respectively, the Newtonian stress tensors in the exterior and interior fluids, $\boldsymbol{n}$ is the unit normal directed into the exterior fluid and $\lambda = \hat{\mu}/\mu$ is the viscosity ratio. The first and third boundary conditions correspond to the continuity of velocity and tangential stresses across the interface, the latter being the case for a surfactant-free drop. Equation (2.6) is the kinematic boundary condition for a steady interface. Finally, we require that the exterior velocity field $\boldsymbol{u}$ approach the ambient linear field, $\boldsymbol{u}^{\infty} = \boldsymbol{\Gamma}\cdot\boldsymbol{r}$, at large distances from the drop, and that the interior velocity field $\hat{\boldsymbol{u}}$ remain finite at the origin. Here, $\boldsymbol{\Gamma}$ denotes the transpose of the (dimensionless) velocity gradient tensor of the ambient linear flow; for simple shear flow, for instance, $\boldsymbol{\Gamma} = \mathbf{1}_x\mathbf{1}_y$, x, y and z being the flow, gradient and vorticity directions. In assuming a specified (spherical) shape for the imposition of the interfacial boundary conditions, one loses the freedom of satisfying, in addition, the normal stress balance at the interface. One may nevertheless determine the correct leading-order velocity and pressure fields using (2.5)–(2.7) in the limit of small Ca, where $Ca = \mu a\dot{\gamma}/T$, T being the coefficient of interfacial tension, is the capillary number; the limit $Ca \ll 1$, therefore, corresponds to the situation in which surface tension is dominant. The normal stress balance,

$$\boldsymbol{\sigma}:\boldsymbol{nn} - \lambda\,\hat{\boldsymbol{\sigma}}:\boldsymbol{nn} = \frac{2}{Ca}\nabla\cdot\boldsymbol{n}, \tag{2.8}$$

may then be used to determine the $O(Ca)$ deviation of the drop from sphericity on account of the imposed external flow.

The above solution procedure may be carried forward to higher orders. For instance, with $Re = 0$, the Stokes velocity fields may be determined at leading order by considering a spherical drop. Equation (2.8) may be used thereafter to determine the $O(Ca)$ drop deformation. The original boundary conditions (2.5)–(2.7) may now be used, together with the $O(Ca)$ deformed interface, in order to determine the $O(Ca)$ corrections to the leading-order Stokes fields. These corrections may in turn be used in (2.8) to calculate the $O(Ca^2)$ deformation, and so on. The protocol remains unaltered in presence of inertia. Thus, the calculation of the velocity and pressure fields to $O(Re)$ does not require one to know the $O(ReCa)$ deformation of the drop, and one may continue to impose the interfacial boundary conditions at a spherical interface even in the limit of small but finite Re.

It is well known that the limit of weak inertia is a singular one (e.g. see Proudman & Pearson 1957; Lin *et al.* 1970). Thus, the leading-order exterior Stokes velocity field obtained from solving the system (2.1)–(2.4) with $Re = \hat{Re} = 0$ is not a uniformly valid approximation in an unbounded domain at any finite Re however small. The approximation breaks down for distances from the drop larger than an inertial screening length that, for an ambient linear flow field, scales as $O(aRe^{-1/2})$. The correct approximation to the velocity field in this outer region must be obtained from a solution of the linearized Navier–Stokes equations instead. However, it is shown in the next section that the non-uniformity of the Stokes approximation, and the resulting modified velocity field in the outer region only affects the bulk stress in a dilute emulsion at $O(\phi Re^{3/2})$. Thus, when determining the bulk stress to $O(\phi Re)$, one may solve for the $O(Re)$ inertial correction to the leading-order Stokes exterior velocity field using a regular perturbation expansion.

Accordingly, we expand both the interior and the exterior velocity fields as

$$\boldsymbol{u} = \boldsymbol{u}^{(0)} + Re\ \boldsymbol{u}^{(1)} + O(Re^{3/2}), \tag{2.9}$$

$$\hat{\boldsymbol{u}} = \hat{\boldsymbol{u}}^{(0)} + Re\ \hat{\boldsymbol{u}}^{(1)} + O(Re^{3/2}), \tag{2.10}$$

with similar expansions for the corresponding pressure fields, recognizing that the singular character of the perturbation only enters at $O(Re^{3/2})$. The leading-order terms in (2.9) and (2.10), of course, satisfy the Stokes equations, and have been well documented for an ambient linear flow (see Leal 1992). We have

$$\boldsymbol{u}^{(0)} = \boldsymbol{\Gamma}\cdot\boldsymbol{r} - \frac{\lambda}{(\lambda+1)r^5}\boldsymbol{E}\cdot\boldsymbol{r} - \left(\frac{(5\lambda+2)}{2(\lambda+1)r^5} - \frac{5\lambda}{2(\lambda+1)r^7}\right)\boldsymbol{r}(\boldsymbol{E}:\boldsymbol{rr}), \tag{2.11}$$

$$p^{(0)} = -\frac{(5\lambda+2)}{(\lambda+1)r^5}\boldsymbol{E}:\boldsymbol{rr}, \tag{2.12}$$

$$\hat{\boldsymbol{u}}^{(0)} = \boldsymbol{\Omega}\cdot\boldsymbol{r} + \left(-\frac{3}{2(\lambda+1)} + \frac{5r^2}{2(\lambda+1)}\right)\boldsymbol{E}\cdot\boldsymbol{r} - \frac{1}{(\lambda+1)}\boldsymbol{r}(\boldsymbol{E}:\boldsymbol{rr}), \tag{2.13}$$

$$\hat{p}^{(0)} = \frac{21\lambda}{2(\lambda+1)}\boldsymbol{E}:\boldsymbol{rr}, \tag{2.14}$$

where $\boldsymbol{E} = (1/2)(\boldsymbol{\Gamma} + \boldsymbol{\Gamma}^{\dagger})$ and $\boldsymbol{\Omega} = (1/2)(\boldsymbol{\Gamma} - \boldsymbol{\Gamma}^{\dagger})$ are the rate of strain tensor and the transpose of the vorticity tensor, respectively, of the ambient linear flow.

At O(Re), one obtains

$$\nabla^2\boldsymbol{u}^{(1)} - \nabla p^{(1)} = \boldsymbol{u}^{(0)}\cdot\nabla\boldsymbol{u}^{(0)}, \tag{2.15}$$

$$\nabla\cdot\boldsymbol{u}^{(1)} = 0, \tag{2.16}$$

and

$$\nabla^2\hat{\boldsymbol{u}}^{(1)} - \nabla\hat{p}^{(1)} = \frac{1}{\lambda}\hat{\boldsymbol{u}}^{(0)}\cdot\nabla\hat{\boldsymbol{u}}^{(0)}, \tag{2.17}$$

$$\nabla\cdot\hat{\boldsymbol{u}}^{(1)} = 0, \tag{2.18}$$

where we have used $\hat{Re} = Re/\lambda$, and $\boldsymbol{u}^{(0)}$ and $\hat{\boldsymbol{u}}^{(0)}$ are given by (2.11) and (2.13), respectively. One again has the boundary conditions (2.5)–(2.7) at $r = 1$, but now written in terms of $\boldsymbol{u}^{(1)}$, $\hat{\boldsymbol{u}}^{(1)}$, $\boldsymbol{\sigma}^{(1)}$ and $\hat{\boldsymbol{\sigma}}^{(1)}$.

The system of equations comprising (2.15)–(2.18) has, in fact, been examined before by Peery (1966). In attempting to verify his calculations, however, we found errors in the expressions for both the interior and exterior velocity fields. The former, in particular, had serious errors. The rather tedious nature of the calculation involved prompted us to directly determine the bulk stress via an alternate approach that does not entail calculation of the $O(Re)$ disturbance fields; this approach involving a reciprocal theorem formulation is detailed in § 4. Herein, we only quote the final results of the regular perturbation analysis for the $O(Re)$ interior and exterior velocity and pressure fields, obtained using *Mathematica* (a symbolic algebra package). These will be used in the next section for the determination of the bulk stress. The solution procedure at $O(Re)$, in fact, closely resembles the calculation of the non-Newtonian correction to the leading-order Stokes velocity field for a neutrally buoyant particle in a second-order fluid undergoing a linear flow, since both the velocity and pressure fields in both cases are quadratic functionals of the ambient velocity gradient tensor. Details of the latter calculation may be found in Koch & Subramanian (2006).

The $O(Re)$ exterior velocity and pressure fields are given by

$$\begin{aligned}
\boldsymbol{u}^{(1)} = \frac{1}{(\lambda+1)}\Bigg[&\left(-\frac{c_1}{4r^{11}}+\frac{c_2}{2r^{10}}-\frac{7c_3}{4r^9}+\frac{c_4}{3r^8}-\frac{c_5}{12r^5}\right)(\boldsymbol{\Gamma}:\boldsymbol{rr})^2\boldsymbol{r}+\left(\frac{c_1}{18r^9}-\frac{3c_2}{32r^8}\right.\\
&\left.+\frac{c_6}{r^7}-\frac{c_4}{36r^6}-\frac{c_7}{2r^5}+\frac{c_5}{18r^3}\right)(\boldsymbol{\Gamma}:\boldsymbol{rr})(\boldsymbol{\Gamma}\cdot\boldsymbol{r})+\left(\frac{c_1}{18r^9}-\frac{3c_2}{32r^8}+\frac{c_8}{r^7}-\frac{c_4}{36r^6}+\frac{c_7}{2r^5}\right.\\
&\left.-\frac{c_5}{9r^3}\right)(\boldsymbol{\Gamma}:\boldsymbol{rr})(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})+\left(\frac{c_1}{36r^9}-\frac{3c_2}{32r^8}+\frac{c_9}{r^7}-\frac{5c_4}{36r^6}-\frac{c_{10}}{r^5}+\frac{c_5}{36r^3}\right)(\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}\cdot\boldsymbol{r})\boldsymbol{r}\\
&+\left(\frac{c_1}{18r^9}-\frac{3c_2}{16r^8}+\frac{c_9+c_{11}}{r^7}-\frac{5c_4}{18r^6}+\frac{c_5}{18r^3}\right)(\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})\boldsymbol{r}+\left(\frac{c_1}{36r^9}-\frac{3c_2}{32r^8}\right.\\
&\left.+\frac{c_{11}}{r^7}-\frac{5c_4}{36r^6}+\frac{c_{10}}{r^5}+\frac{c_5}{36r^3}\right)(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})\boldsymbol{r}+\left(-\frac{c_1}{126r^7}+\frac{c_2}{32r^6}-\frac{c_{17}}{r^5}+\frac{c_4}{36r^4}\right.\\
&\left.-\frac{\lambda+1}{30r^3}+\frac{c_5}{9r}\right)\boldsymbol{\Gamma}\cdot(\boldsymbol{\Gamma}\cdot\boldsymbol{r})+\left(-\frac{c_1}{126r^7}+\frac{c_2}{32r^6}-\frac{c_{18}}{r^5}+\frac{c_4}{36r^4}+\frac{\lambda+1}{30r^3}-\frac{c_5}{18r}\right)\boldsymbol{\Gamma}^\dagger\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})\\
&+\left(-\frac{c_1}{126r^7}+\frac{c_2}{32r^6}-\frac{c_{12}}{r^5}+\frac{c_4}{36r^4}-\frac{c_7}{6r^3}+\frac{c_5}{9r}\right)\boldsymbol{\Gamma}^\dagger\cdot(\boldsymbol{\Gamma}\cdot\boldsymbol{r})+\left(-\frac{c_1}{126r^7}+\frac{c_2}{32r^6}-\frac{c_{13}}{r^5}\right.\\
&\left.+\frac{c_4}{36r^4}+\frac{c_7}{6r^3}-\frac{c_5}{18r}\right)\boldsymbol{\Gamma}\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})+\left(-\frac{c_1}{252r^7}+\frac{c_2}{48r^6}-\frac{c_{12}+c_{13}}{2r^5}+\frac{c_4}{18r^4}-\frac{c_5}{36r}\right)\\
&(\boldsymbol{\Gamma}:\boldsymbol{\Gamma}^\dagger+\boldsymbol{\Gamma}:\boldsymbol{\Gamma})\boldsymbol{r}\Bigg],
\end{aligned} \tag{2.19}$$

$$\begin{aligned}
p^{(1)} = -\frac{1}{2}(\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})+\frac{1}{(\lambda+1)}\Bigg[&\left(-\frac{5c_{14}}{2r^{12}}+\frac{c_2}{r^{10}}-\frac{7c_3}{4r^9}+\frac{3c_4}{4r^8}-\frac{5c_7}{r^7}+\frac{c_5}{2r^5}\right)(\boldsymbol{\Gamma}:\boldsymbol{rr})^2\\
&+\left(-\frac{c_{14}}{2r^{10}}+\frac{c_3}{2r^7}-\frac{c_4}{4r^6}-\frac{c_{15}}{r^5}-\frac{c_5}{6r^3}\right)(\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}\cdot\boldsymbol{r})+\left(-\frac{c_{14}}{r^{10}}+\frac{c_3}{r^7}-\frac{c_4}{2r^6}+\frac{c_7}{r^5}\right)\\
&(\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})+\left(-\frac{c_{14}}{2r^{10}}+\frac{c_3}{2r^7}-\frac{c_4}{4r^6}+\frac{2c_{10}}{r^5}+\frac{c_5}{6r^3}\right)(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})\\
&+\left(-\frac{c_3}{10r^5}+\frac{c_4}{12r^4}-\frac{c_5}{6r}\right)(\boldsymbol{\Gamma}:\boldsymbol{\Gamma}^\dagger+\boldsymbol{\Gamma}:\boldsymbol{\Gamma})\Bigg].
\end{aligned} \tag{2.20}$$

The $O(Re)$ interior velocity and pressure fields are given by

$$\hat{\boldsymbol{u}}^{(1)} = \frac{1}{\lambda(\lambda+1)^2}\Bigg[\left(c_1'r^2 + c_2'\right)(\boldsymbol{\Gamma}:\boldsymbol{rr})^2\boldsymbol{r} + \left(-\frac{9c_1'r^4}{8} - c_3'r^2 + c_4'\right)(\boldsymbol{\Gamma}:\boldsymbol{rr})(\boldsymbol{\Gamma}\cdot\boldsymbol{r})$$
$$+\left(-\frac{9c_1'r^4}{8} - c_5'r^2 - c_6'\right)(\boldsymbol{\Gamma}:\boldsymbol{rr})(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r}) + \left(-\frac{5c_1'r^4}{6} + c_7'r^2 - c_8'\right)(\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}\cdot\boldsymbol{r})\boldsymbol{r}$$
$$+\left(-\frac{5c_1'r^4}{3} + 2c_7'r^2 - (c_8' + c_9')\right)(\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})\boldsymbol{r} + \left(-\frac{5c_1'r^4}{6} + c_7'r^2 - c_9'\right)$$
$$\times(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})\boldsymbol{r} + \left(\frac{23c_1'r^6}{16} + c_{25}'r^4 + c_{27}'r^2 + c_{29}'\right)\boldsymbol{\Gamma}\cdot(\boldsymbol{\Gamma}\cdot\boldsymbol{r}) + \left(\frac{23c_1'r^6}{16}\right.$$
$$\left. + c_{26}'r^4 + c_{28}'r^2 + c_{30}'\right)\boldsymbol{\Gamma}^\dagger\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r}) + \left(\frac{23c_1'r^6}{16} + c_{10}'r^4 + c_{11}'r^2 - c_{12}'\right)\boldsymbol{\Gamma}^\dagger\cdot(\boldsymbol{\Gamma}\cdot\boldsymbol{r})$$
$$+\left(\frac{23c_1'r^6}{16} + c_{13}'r^4 + c_{14}'r^2 - c_{15}'\right)\boldsymbol{\Gamma}\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r}) + \left(-\frac{23c_1'r^6}{72} - \frac{(c_{10}' + c_{13}')}{7}r^4\right.$$
$$\left. - \frac{(c_{11}' + c_{14}')}{5}r^2 + \frac{(c_{12}' + c_{15}')}{3}\right)(\boldsymbol{\Gamma}:\boldsymbol{\Gamma}^\dagger + \boldsymbol{\Gamma}:\boldsymbol{\Gamma})\boldsymbol{r}\Bigg], \tag{2.21}$$

$$\hat{p}^{(1)} = \frac{1}{\lambda(\lambda+1)^2}\Bigg[\left(-13c_{16}'r^2 - c_{17}'\right)(\boldsymbol{\Gamma}:\boldsymbol{rr})^2 + \left(\frac{53c_1'r^4}{48} + c_{18}'r^2 + c_{19}'\right)(\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}\cdot\boldsymbol{r})$$
$$+\left(\frac{53c_1'r^4}{24} + (c_{18}' + c_{19}')r^2 + c_{21}'\right)(\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r}) + \left(\frac{53c_1'r^4}{48} + c_{20}'r^2 + c_{22}'\right)$$
$$(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r}) + \left(-\frac{133c_1'r^6}{144} - c_{23}'r^4 - c_{24}'r^2\right)(\boldsymbol{\Gamma}:\boldsymbol{\Gamma}^\dagger + \boldsymbol{\Gamma}:\boldsymbol{\Gamma})\Bigg]. \tag{2.22}$$

The constants c_i and c_i', appearing in the above expressions, have been tabulated in Appendix A. We have verified that the expression (2.19) for the exterior velocity field reduces to that for a rigid particle in the limit $\lambda \to \infty$. The latter velocity field has been correctly determined by Peery (1966) for a planar linear flow (also see Subramanian & Koch 2006). In the same limit, all constants in the interior velocity field except c_{29}' and c_{30}' vanish, and the interior velocity field reduces to $\lim_{\lambda\to\infty}\hat{\boldsymbol{u}}^{(1)} = (1/6)\,[\boldsymbol{\Gamma}\cdot(\boldsymbol{\Gamma}\cdot\boldsymbol{r}) - \boldsymbol{\Gamma}^\dagger\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{r})] = (1/6)(\boldsymbol{\omega}\cdot\boldsymbol{E})\wedge\boldsymbol{r}$, indicative of a modification of the angular velocity of a particle, at $O(Re)$, proportional to $\boldsymbol{\omega}\cdot\boldsymbol{E}$, $\boldsymbol{\omega}$ being the ambient vorticity (the actual $O(Re)$ correction to the angular velocity of a torque-free neutrally buoyant particle is not $(Re/6)(\boldsymbol{\omega}\cdot\boldsymbol{E})$, however; there arises an additional inertial contribution, $-(Re/30)(\boldsymbol{\omega}\cdot\boldsymbol{E})$, from the angular acceleration in the ambient linear flow, and the total angular velocity is therefore given by $\Omega_p = (1/2)\,\boldsymbol{\omega} + (2Re/15)\,(\boldsymbol{\omega}\cdot\boldsymbol{E})$, to $O(Re)$, in a general linear flow (Stone *et al.* 2000). The above findings are consistent with the earlier results of Stone *et al.* (2000), who found the angular velocity of a solid particle to remain unchanged to $O(Re)$ in ambient linear flows without vortex stretching ($\boldsymbol{\omega}\cdot\boldsymbol{E} = 0$); in simple shear, for instance, the first correction to the particle angular velocity arises at $O(Re^{3/2})$ (Lin *et al.* 1970).

As indicated above, a uniformly valid solution of the original system of equations, (2.1)–(2.4), for small Re, requires a matched asymptotic expansions approach. We therefore expect that the inertial correction, $\boldsymbol{u}^{(1)}$, will not satisfy the far-field boundary condition. This is, in fact, easily seen from (2.19); the leading-order terms in $\boldsymbol{u}^{(1)}$ remain $O(1)$ in the limit $r \gg 1$. The non-uniform nature of the perturbation expansion may also be seen from a simple scaling argument. Since the far-field

Stokes velocity disturbance is that due to a stresslet, $\boldsymbol{u}^{(0)} - \boldsymbol{\Gamma}\cdot\boldsymbol{r} \sim O(1/r^2)$ for $r \gg 1$, $\boldsymbol{u}^{(0)}\cdot\nabla\boldsymbol{u}^{(0)} \sim (\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot\nabla[\boldsymbol{u}^{(0)} - \boldsymbol{\Gamma}\cdot\boldsymbol{r}] \sim O(1/r^2)$; using (2.15), this in turn implies $\boldsymbol{u}^{(1)}$ is independent of r for large r as above, and thus fails to satisfy the far-field decay condition. In the context of the present calculation, one only needs to recognize that the expression for $\boldsymbol{u}^{(1)}$ above still remains valid for $r \ll Re^{-1/2}$, and this allows one to calculate the bulk stress to $O(\phi Re)$.

Furthermore, although it appears as if the expressions for $\boldsymbol{u}^{(1)}$ and $p^{(1)}$ are valid for an arbitrary $\boldsymbol{\Gamma}$, this is not so. The calculation of the disturbance velocity and pressure fields, and thence the stresslet, associated with a particle or a drop immersed in an arbitrary imposed linear flow field becomes a rather difficult one at finite Re. While the Stokes equations admit an arbitrary linear flow as a possible solution, the associated pressure field being a constant, this is not the case for the Navier–Stokes equations. The only linear flows that remain exact solutions of the Navier–Stokes equations are extensional flow ($\boldsymbol{\Gamma} = \boldsymbol{E}$) and the one-parameter family of planar linear flows ($\boldsymbol{\omega}\cdot\boldsymbol{E} = 0$; the latter include both simple shear flow and solid-body rotation ($\boldsymbol{\Gamma} = -\boldsymbol{\Gamma}^{\dagger}$) (Subramanian & Koch 2006). Except for the degenerate instance of simple shear flow, there is a non-trivial quadratic pressure field at $O(Re)$ in each of these cases; this is represented by the first term in (2.20). The restriction on the nature of the ambient linear flow may be readily understood from the vorticity equation

$$Re\left(\frac{\mathrm{D}\boldsymbol{\omega}}{\mathrm{D}t} - \boldsymbol{\omega}\cdot\boldsymbol{E}\right) = \nabla^2\boldsymbol{\omega}, \tag{2.23}$$

where the vortex-stretching term is seen to be absent only in the aforementioned exceptional cases, thereby allowing for a solution with a constant vorticity field. Steady linear flows with vortex stretching do not satisfy the Navier–Stokes equations. Thus, for small but finite Re, consideration of a general three-dimensional linear flow entails an inconsistency at $O(Re)$ arising from such a flow not being an exact solution of the governing equations to the required order. The issue of deriving a disturbance velocity field accurate to $O(Re)$ becomes a rather delicate one, since one must also show that the contributions to the disturbance velocity and pressure fields arising from additional cubic or unsteady terms in the ambient flow (required for it to exactly solve the Navier–Stokes equations to $O(Re)$) are, in an appropriate sense, smaller than those arising from the linear component, and included in (2.19) and (2.20). This aspect has been looked into in the context of an inertial suspension (Stone *et al.* 2000), and we examine it in more detail for an emulsion in § 4, where we calculate the bulk stress via a reciprocal theorem formulation. The latter approach does not require one to solve for the $O(Re)$ disturbance fields and is, therefore, more convenient for the bulk stress determination in an arbitrary linear flow. For now, we only note that the above inconsistency being absent for linear flows without vortex stretching, the expressions (2.19) and (2.20) remain valid for this class of flows. In the next section, we determine the exterior stress field from (2.19) and (2.20), and use it to calculate the bulk stress, to $O(\phi Re)$, in a dilute emulsion subjected to a three-dimensional extensional flow or a planar linear flow.

3. Bulk stress in a dilute emulsion: linear flows without vortex stretching

In this section, we determine the bulk stress to $O(\phi Re)$ in a dilute emulsion subject to a linear flow without vortex stretching, that is, an ambient linear flow that itself remains an exact solution of the Navier–Stokes equations for any Re. An important example of such a flow, particularly from the rheological perspective, is simple shear flow, and the results of the analysis here will be used in § 5 to examine the behaviour of the normal stress differences of a dilute emulsion in simple shear.

The expression for the bulk stress in a dispersion of force-free drops is given by

$$\Sigma_{ij} = -p_t\delta_{ij} + 2\mu E_{ij} + \Sigma_{ij}^{(d)}, \tag{3.1}$$

where p_t is an arbitrary isotropic pressure, the second term is the deviatoric stress in the Newtonian suspending fluid, and $\Sigma_{ij}^{(d)}$ is the excess stress because of the disperse phase, defined in terms of a volume average (see Batchelor 1970):

$$\Sigma_{ij}^{(d)} = \frac{1}{V}\sum_{\alpha=1}^{N}\int_{A_\alpha^+}\left[\frac{1}{2}(\sigma_{ik}r_j + \sigma_{jk}r_i)n_k - \mu(u_i n_j + u_j n_i)\right]\mathrm{d}A$$
$$- \frac{1}{V}\sum_{\alpha=1}^{N}\int_{V_\alpha}\frac{1}{2}(\rho f_i' r_j + \rho f_j' r_i)\mathrm{d}V - \frac{1}{V}\int_V \rho\, u_i' u_j'\,\mathrm{d}V. \tag{3.2}$$

Here, V_α and A_α denote the volume and surface area of the αth drop and the summations run over all the N drops in the microscopic volume V; the surface A_α^+ includes the thin layer, acted on by interfacial tension forces, around each drop. Furthermore, f_i' denotes the local acceleration relative to the average, and u_i' is the local fluctuation velocity relative to the ambient linear flow; thus, $u_i' = u_i - \Gamma_{ij}r_j$ for $r > 1$, and $u_i' = \hat{u}_i - \Gamma_{ij}r_j$ for $r < 1$, with r denoting radial distance from the centre of any given drop. The first term in (3.2) is the familiar stresslet that leads to the Einstein correction for a dilute suspension of rigid particles for $Re = 0$ (Kim & Karrila 1991); for an emulsion, it is equal to the sum of the viscous and surface tension stresses. The second and third terms are the acceleration stress and the Reynolds stress, respectively, and are relevant only for finite Re (Batchelor 1970). The averaging procedure used to define the bulk stress conceals velocity fluctuations (relative to the imposed mean flow) at the microscale. Thus, as is the case in turbulence, the acceleration and Reynolds stresses represent the processes of momentum transfer across a given surface on account of these microscopic velocity fluctuations. Strictly speaking, E_{ij} in (3.1) is the average rate of strain in the emulsion, and must be distinguished from the rate of strain in the suspending fluid alone (as defined, for instance, in § 2 via a far-field boundary condition). The non-affine nature of the drop motion implies that the two rates of strain need not be the same, and in particular, for the case of very viscous drops (or rigid particles), the rate of strain in the suspending fluid must actually be higher in order to conform to the imposed average value. Thus, the far-field rate of strain encountered in § 2 is interpreted now as that corresponding to the ambient linear flow of the emulsion.

For the case of a dilute emulsion of non-interacting drops, the summations in (3.2) comprise N identical terms. Using the scalings introduced in the previous section and the disperse phase volume fraction, $\phi = \{N[(4/3)\pi a^3]\}/V$, (3.2) reduces to

$$\Sigma_{ij}^{(d)} = \frac{3\phi}{4\pi}\int_{A_d^+}\left[\frac{1}{2}(\sigma_{ik}r_j + \sigma_{jk}r_i)n_k - (u_i n_j + u_j n_i)\right]\mathrm{d}A$$
$$- \frac{3\phi}{4\pi}Re\int_{V_d}\frac{1}{2}(f_i' r_j + f_j' r_i)\,\mathrm{d}V - \frac{3\phi}{4\pi}Re\int_V u_i' u_j'\,\mathrm{d}V, \tag{3.3}$$

where V_d and A_d^+ now denote the volume and surface area of an isolated neutrally buoyant drop in an unbounded fluid domain V undergoing a planar linear flow.

The velocity fields and the corresponding stress fields, to be used in (3.3), have been obtained in § 2.

The distinction in the particular surface of integration (external to or within the interfacial layer) to be used in the calculation of the stresslet is an important one when calculating the emulsion stress, since the normal component of the stress has a discontinuity at the interface arising from surface tension. As emphasized by Batchelor (see Batchelor 1970), the above expression for the bulk stress is also valid for drops provided one interprets the surface area of integration in the stresslet term as one that includes the interfacial layer. This then resolves the ambiguity with regard to the choice of the surface force density to be used in the stresslet integral; the stresses to be used in this integral correspond to those in the exterior fluid in the limit $r \to 1$. One may similarly show that the singular surface tension stress does not contribute to either the acceleration or Reynolds stress integrals, although the domain of integration in both cases includes the interfacial layer (see Batchelor 1970). The integrations in (3.3) may be carried out assuming the surface of the drop to be that of a sphere. The contribution to the viscosity, at leading order, that arises, for instance, from the interfacial tension forces acting within the interface of a deformed drop is automatically accounted for.

The separate contributions to the bulk stress due to viscous and interfacial tension forces may be seen explicitly from an alternate expression for the bulk stress, which may be derived from (3.3) as follows. Applying the divergence theorem to the terms involving the force density in (3.3), one obtains

$$\Sigma_{ij}^{(d)} = \frac{3\phi}{4\pi}\left[\int_{V_d}\frac{1}{2}\left(\frac{\partial\sigma_{ik}}{\partial r_k}r_j + \frac{\partial\sigma_{jk}}{\partial r_k}r_i\right)\mathrm{d}V + \int_{V_d^+}\sigma_{ij}\mathrm{d}V\right] - \frac{3\phi}{4\pi}\int_{A_d}(u_i n_j + u_j n_i)\,\mathrm{d}A - \frac{3\phi}{4\pi}Re\int_{V_d}\frac{1}{2}(f_i' r_j + f_j' r_i)\mathrm{d}V - \frac{3\phi}{4\pi}Re\int_V u_i' u_j'\,\mathrm{d}V, \quad (3.4)$$

where both the volume integral involving the stress divergence and the surface integral involving the velocity field are continuous across the interface (the superscripts $\pm$ being redundant). Now, we use $\nabla\cdot\boldsymbol{\sigma} = Re\,\boldsymbol{f}$ to eliminate the acceleration stress integral and split the integral of $\boldsymbol{\sigma}$ over V_d^+ into one over V_d^- and one involving the singular interfacial stresses. The latter term may be expressed as an integral over the drop interface, and one obtains

$$\Sigma_{ij}^{(d)} = \frac{3\phi}{4\pi}(\lambda - 1)\int_{A_d}(u_i n_j + u_j n_i)\,\mathrm{d}A + \frac{3\phi}{4\pi Ca}\int_{V_d}(\delta_{ij} - n_i n_j)\,\mathrm{d}A - \frac{3\phi}{4\pi}Re\int_V u_i' u_j'\,\mathrm{d}V. \quad (3.5)$$

Note that the leading-order contribution in (3.5) arises from the $O(1/Ca)$ surface tension term, but this is isotropic, and therefore, of no relevance to the present rheological calculation. It is, however, clear that an $O(Ca)$ shape deformation in the surface tension integral will indeed lead to an $O(1)$ viscous contribution and, therefore, one needs to account for the deviation of the drop from sphericity when using (3.5) for the stress calculation. In fact, with $Re = 0$, and for drops of the same viscosity ($\lambda = 1$), the enhancement in the viscosity of a dilute emulsion over that of the suspending fluid comes only from the existence of an interfacial tension. This contribution is accounted for in Taylor's original calculation of the emulsion viscosity, $\mu_{eff} = \mu[1 + \phi\{(5\lambda + 2)/(2(\lambda + 1))\}]$, where with $\lambda = 1$, one obtains $\mu_{eff} = \mu[1 + \{(7/4)\phi\}]$ (see Schowalter *et al.* 1968).

Returning to the original expression (3.3) for the stress, one may simplify the inertial contributions by using an explicit expression for the acceleration f_i'. For the special case of simple shear flow, there being no ambient acceleration, one has $f_i = f_i' = (D\hat{u}_i)/(Dt)$. Even for the case of a general linear flow, however, a neutrally buoyant drop in the limit $Ca \ll 1$ translates, to $O(Re)$, with the velocity of the ambient flow at its centre, and the only additional acceleration in the drop phase is therefore that due to the interior velocity field in a reference frame that translates with the drop; in other words, $f_i' = (D\hat{u}_i)/(Dt)$, at leading order, even for a general linear flow. Using this, the acceleration stress in (3.3) becomes

$$\left.\begin{aligned} \frac{1}{2}\int_{V_d}\left(f_i' r_j + f_j' r_i\right) dV &= \frac{1}{2}\int_{V_d}\left(\frac{D\hat{u}_i}{Dt} r_j + \frac{D\hat{u}_j}{Dt} r_i\right) dV, \\ &= \frac{1}{2}\int_{V_d}\left(\hat{u}_l \frac{\partial \hat{u}_i}{\partial r_l} r_j + \hat{u}_l \frac{\partial \hat{u}_j}{\partial r_l} r_i\right) dV, \\ &= -\int_{V_d} \hat{u}_i \hat{u}_j \, dV, \end{aligned}\right\} \tag{3.6}$$

where we have used the divergence theorem, and the fact that $\hat{\boldsymbol{u}} \cdot \boldsymbol{n} = 0$ at the interface. Using this simplified expression for the acceleration stress in (3.3), one obtains

$$\Sigma_{ij}^{(d)} = \frac{3\phi}{4\pi}\left[\int_{A_d^+}\left[\frac{1}{2}(\sigma_{ik} r_j + \sigma_{jk} r_i) n_k - (u_i n_j + u_j n_i)\right] dA \right. \\ \left. - Re\left(-\int_{V_d} \hat{u}_i \hat{u}_j \, dV + \int_{V_d} u_i' u_j' \, dV + \int_{V-V_d} u_i' u_j' \, dV\right)\right], \tag{3.7}$$

where we have now divided the Reynolds stress into interior (V_d) and exterior contributions $(V - V_d)$. Since $u_i' = \hat{u}_i - \Gamma_{ij} r_j$ when $r < 1$, (3.7) may be further written in the form

$$\Sigma_{ij}^{(d)} = \frac{3\phi}{4\pi}\left[\int_{A_d^+}\left[\frac{1}{2}(\sigma_{ik} r_j + \sigma_{jk} r_i) n_k - (u_i n_j + u_j n_i)\right] dA \right. \\ \left. + Re\left(\frac{4\pi}{15}\Gamma_{ik}\Gamma_{jk} + \int_{V_d} \Gamma_{ik} r_k u_j' \, dV + \int_{V_d} \Gamma_{jk} r_k u_i' \, dV - \int_{V-V_d} u_i' u_j' \, dV\right)\right]. \tag{3.8}$$

The most tedious part of the stress calculation using (3.8) is the determination of the $O(Re)$ correction to the stresslet. It is this contribution alone that necessitated the calculation of the $O(Re)$ corrections to the interior and exterior Stokes fields in § 2. The contributions in (3.8) arising from the acceleration and Reynolds stress terms in (3.3) are already $O(Re)$ and, therefore, a calculation accurate to $O(Re)$ need to use only the leading-order Stokes fields. We also note that since the Stokes disturbance velocity field is $O(1/r^2)$ for $r \gg 1$, the integrand of the last term in (3.8) is $O(1/r^4)$ for $r \gg 1$, and the leading-order Reynolds stress integral over the unbounded fluid domain, $\int_{V-V_d} u_i'^{(0)} u_j'^{(0)} dV$, is therefore convergent. Carrying this argument further, we note that the Stokes approximation breaks down for $r \geqslant O(Re^{-1/2})$. Assuming that the modified outer velocity field decays at least as rapidly, the resulting modification to the stress must scale as $O(\phi Re). \int_{O(Re^{-1/2})}^{\infty} O(1/r^4) dV \sim O(\phi Re^{3/2})$ (this assumption relies on the intuitive notion that convection is a more efficient mode of momentum transfer at length scales larger than the inertial screening length and outside of a wake of an asymptotically small extent. This is borne out for the simpler problem of a particle translating in an otherwise quiescent fluid; Leal 1992). One may arrive at the same conclusion by instead considering the corrections to the leading-order Reynolds stress contribution in the exterior fluid domain involving $\boldsymbol{u}^{(1)}$. This is given by $Re^2 \int [u'^{(0)}_i u'^{(1)}_j + u'^{(1)}_i u'^{(0)}_j] \mathrm{d}V + Re^3 \int u'^{(1)}_i u'^{(1)}_j \mathrm{d}V$. Since $\boldsymbol{u}^{(1)}$ remains $O(1)$ for $r \gg 1$ (see § 2), both integrals are evidently divergent. This divergence is cutoff at the inertial screening length of $O(Re^{-1/2})$, again indicating that the non-uniformity of the leading-order Stokes approximation affects the stress calculation only at $O(\phi Re^{3/2})$.

In accordance with our expansions for the velocity and pressure fields for small Re, viz. (2.9) and (2.10), we now split the stress given by (3.8) into an $O(1)$ and an $O(Re)$ contribution. Thus, using $\boldsymbol{\sigma} = \boldsymbol{\sigma}^{(0)} + Re\, \boldsymbol{\sigma}^{(1)}$, one obtains

$$\Sigma^{(d)}_{ij} = \Sigma^{(d0)}_{ij} + Re\, \Sigma^{(d1)}_{ij} + O(\phi Re^{3/2}), \tag{3.9}$$

where,

$$\Sigma^{(d0)}_{ij} = \frac{3\phi}{4\pi} \int_{A_d^+} \left[\frac{1}{2} \left(\sigma^{(0)}_{ik} r_j + \sigma^{(0)}_{jk} r_i \right) n_k - \left(u^{(0)}_i n_j + u^{(0)}_j n_i \right) \right] \mathrm{d}A, \tag{3.10}$$

$$\Sigma^{(d1)}_{ij} = \frac{3\phi}{4\pi} \int_{A_d^+} \left[\frac{1}{2} \left(\sigma^{(1)}_{ik} r_j + \sigma^{(1)}_{jk} r_i \right) n_k - \left(u^{(1)}_i n_j + u^{(1)}_j n_i \right) \right] \mathrm{d}A$$
$$+ \frac{3\phi}{4\pi} \left(\frac{4\pi}{15} \Gamma_{ik} \Gamma_{jk} + \int_{V_d} \Gamma_{ik} r_k u'^{(0)}_j \mathrm{d}V + \int_{V_d} \Gamma_{jk} r_k u'^{(0)}_i \mathrm{d}V - \int_{V-V_d} u'^{(0)}_i u'^{(0)}_j \mathrm{d}V \right). \tag{3.11}$$

If the drop is assumed to be spherical, then the surface integrations in (3.10) and (3.11) reduce to those over a unit sphere, and $\Sigma^{(d0)}_{ij}$, in particular, simplifies to a Newtonian form; thus, $\Sigma^{(d0)}_{ij} = [\mu\{(5\lambda + 2)/(\lambda + 1)\}\phi\, E_{ij}] + O(\phi Ca)$ in the limit $Ca \ll 1$. Unlike a dilute suspension, however, a dilute emulsion, even at $Re = 0$, and in the absence of hydrodynamic interactions between drops, does not behave as a Newtonian fluid. The Newtonian term with an effective viscosity given by the Taylor contribution is therefore only the leading-order approximation in the limit of small Ca. Accounting for drop deformation leads to a non-Newtonian rheology at higher orders in Ca. This is hardly surprising since the presence of an interfacial tension gives rise to an intrinsic relaxation time, $\tau_r \sim O(a\mu/T)$, and one expects a deformed microstructure (drop shape) leading to a non-Newtonian rheology when the flow time scale $\dot{\gamma}^{-1}$ is of the same order as τ_r. The ratio $\tau_r/\dot{\gamma}^{-1}$ is, of course, Ca, and the capillary number is then a natural measure of the non-Newtonian character of a dilute emulsion in the inertialess limit ($Re = 0$). The rheology of such an emulsion, to $O(\phi Ca)$, has been calculated long ago (e.g. see Schowalter *et al.* 1968; Frenkel & Acrivos 1970), and one finds the following expression for the bulk stress:

$$\Sigma^{(d0)}_{ij} = \frac{(5\lambda + 2)}{(\lambda + 1)} \phi E_{ij} + \phi Ca \left[-\frac{1}{80} \left(\frac{19\lambda + 16}{\lambda + 1} \right)^2 \omega_s (\epsilon_{ksi} E_{kj} + \epsilon_{ksj} E_{ki}) \right.$$
$$\left. + \frac{3(19\lambda + 16)(25\lambda^2 + 41\lambda + 4)}{140(\lambda + 1)^3} \left\{ E_{ik} E_{kj} - \frac{1}{3} \delta_{ij} (E_{kl} E_{kl}) \right\} \right] + O(\phi^2, \phi Ca^2), \tag{3.12}$$

for a steady linear flow.

The purpose of this paper to evaluate $\Sigma^{(d1)}_{ij}$ in (3.9) and, hence, the $O(\phi Re)$ contributions to the bulk stress for an ambient linear flow. In accordance with (3.11), the calculation may be divided into two parts. The easier part, the direct inertial effect, involves the Reynolds stress integrals and is performed first followed by the evaluation of the stresslet integral. Using the expressions for the Stokes velocity fields $\boldsymbol{u}^{(0)}$ and $\hat{\boldsymbol{u}}^{(0)}$, given by (2.11) and (2.13), and the standard identity for the angular integration of an even-ordered unit normal polyad over the unit sphere (Bird, Armstrong & Hassager 1987), the following results, valid for a general linear flow, are readily obtained:

$$\int_{V_d} \Gamma_{ik} r_k u_j'^{(0)}\, \mathrm{d}V = -\frac{4\pi}{15} E_{jk}\Gamma_{ik}, \tag{3.13}$$

$$\int_{V_d} \Gamma_{jk} r_k u_i'^{(0)}\, \mathrm{d}V = -\frac{4\pi}{15} E_{ik}\Gamma_{jk}, \tag{3.14}$$

$$\int_{V-V_d} u_i'^{(0)} u_j'^{(0)}\, \mathrm{d}V = \left[\frac{8\pi}{105}\frac{(5\lambda+2)^2}{(\lambda+1)^2} - \frac{8\pi}{105}\frac{\lambda(5\lambda+2)}{(\lambda+1)^2} + \frac{4\pi}{35}\frac{\lambda^2}{(\lambda+1)^2}\right] E_{ik}E_{kj}, \tag{3.15}$$

where we have omitted any isotropic part that emerges from the integrations. The contribution of the inertial stresses in (3.11) may therefore be written as

$$\left.\begin{aligned} &\frac{3\phi}{4\pi}\left(\frac{4\pi}{15}\Gamma_{ik}\Gamma_{jk} + \textstyle\int_{V_d} \Gamma_{ik} r_k u_j'^{(0)}\, \mathrm{d}V + \int_{V_d} \Gamma_{jk} r_k u_i'^{(0)}\, \mathrm{d}V - \int_{V-V_d} u_i'^{(0)} u_j'^{(0)}\, \mathrm{d}V\right) \\ &\quad = \phi\left[\frac{1}{5}\Gamma_{ik}\Gamma_{jk} - \frac{1}{5}(E_{jk}\Gamma_{ik} + E_{ik}\Gamma_{jk}) - \left(\frac{2(5\lambda+2)^2}{35(\lambda+1)^2} - \frac{2\lambda(5\lambda+2)}{35(\lambda+1)^2} + \frac{3\lambda^2}{35(\lambda+1)^2}\right)E_{ik}E_{kj}\right], \\ &\quad = \phi\left[\frac{1}{5}\Gamma_{ik}\Gamma_{jk} - \frac{1}{5}(E_{jk}\Gamma_{ik} + E_{ik}\Gamma_{jk}) - \frac{(43\lambda^2+36\lambda+8)}{35(\lambda+1)^2}E_{ik}E_{kj}\right], \end{aligned}\right\} \tag{3.16}$$

where we again note that the above expression is valid for a general linear flow with vortex stretching.

We now evaluate the indirect inertial effect due to a modification of the surface force density at $O(Re)$ and the resulting correction to the stresslet integral, that is, the first term in (3.11) which represents the combined effect of interfacial and the viscous stresses associated with the $O(Re)$ velocity field. The expression for the $O(Re)$ surface force density is quite lengthy and has, therefore, been relegated to Appendix B. As discussed in § 2, the use of (2.19) and (2.20) restricts the validity of this calculation to ambient linear flows where vortex stretching is absent. Carrying out the integrations over the unit sphere, one finally obtains the following expression for the stresslet integral:

$$\begin{aligned} \int_{A_d^+}\left[\frac{1}{2}\left(\sigma_{ik}^{(1)} r_j + \sigma_{jk}^{(1)} r_i\right)n_k - \left(u_i^{(1)} n_j + u_j^{(1)} n_i\right)\right]\mathrm{d}A = \frac{2\pi}{15}(\Gamma_{ik}\Gamma_{kj} + \Gamma_{jk}\Gamma_{ki}) \\ - \frac{16\pi(3\lambda^2+3\lambda+1)}{27(\lambda+1)^2}(\Omega_{ik}E_{jk} + \Omega_{jk}E_{ik}) + \frac{4\pi(43\lambda^2+36\lambda+8)}{105(\lambda+1)^2}E_{ik}E_{jk}, \end{aligned} \tag{3.17}$$

where the first term arises from the $O(Re)$ pressure gradient in the ambient flow (the first term in (2.20)) and is independent of λ; it vanishes for the case of simple shear flow. We have again omitted all isotropic contributions keeping in mind that it is the deviatoric stress that is of interest.

Using (3.12), (3.16) and (3.17), one arrives at the following expression for the excess stress due to the disperse phase in a dilute emulsion subject to a linear flow without vortex stretching:

$$\Sigma_{ij}^{(d)} = \frac{(5\lambda+2)}{(\lambda+1)}\,\phi E_{ij} + (\phi Ca)\Bigg[-\frac{1}{80}\left(\frac{19\lambda+16}{\lambda+1}\right)^2 \omega_s(\epsilon_{ksi}E_{kj}+\epsilon_{ksj}E_{ki}) + \frac{3(19\lambda+16)(25\lambda^2+41\lambda+4)}{140(\lambda+1)^3}\left\{E_{ik}E_{kj}-\frac{1}{3}\delta_{ij}(E_{kl}E_{kl})\right\}\Bigg] + (\phi Re)\Bigg[\frac{1}{5}\left[\Gamma_{ik}\Gamma_{jk}-(E_{jk}\Gamma_{ik}+E_{ik}\Gamma_{jk})\right] + \frac{1}{10}(\Gamma_{ik}\Gamma_{kj}+\Gamma_{jk}\Gamma_{ki}) - \frac{4(3\lambda^2+3\lambda+1)}{9(\lambda+1)^2} \times(\Omega_{ik}E_{jk}+\Omega_{jk}E_{ik})\Bigg] + O(\phi^2, \phi Re^{3/2}, \phi Ca^2). \tag{3.18}$$

The term proportional to $\boldsymbol{E}\cdot\boldsymbol{E}$ in the stresslet (3.17) cancels the last term in the Reynolds stress contribution (3.16), and as a result, the dependence of the bulk stress on λ, at $O(\phi Re)$, is entirely contained in terms proportional to $\boldsymbol{\Omega}\cdot\boldsymbol{E}$ or $\boldsymbol{E}\cdot\boldsymbol{\Omega}$. Furthermore, the terms independent of λ that remain in (3.18) also cancel, and one obtains the following much simpler form for $\Sigma_{ij}^{(d)}$:

$$\Sigma_{ij}^{(d)} = \frac{(5\lambda+2)}{(\lambda+1)}\,\phi E_{ij} + (\phi Ca)\Bigg[\frac{1}{80}\left(\frac{19\lambda+16}{\lambda+1}\right)^2(\Omega_{ik}E_{jk}+\Omega_{jk}E_{ik}) + \frac{3(19\lambda+16)(25\lambda^2+41\lambda+4)}{140(\lambda+1)^3}E_{ik}E_{jk} - \frac{(19\lambda+16)(25\lambda^2+41\lambda+4)}{140(\lambda+1)^3}(E_{kl}E_{kl})\delta_{ij}\Bigg] - (\phi Re)\frac{4(3\lambda^2+3\lambda+1)}{9(\lambda+1)^2}(\Omega_{ik}E_{jk}+\Omega_{jk}E_{ik}) + O(\phi^2, \phi Re^{3/2}, \phi Ca^2), \tag{3.19}$$

for an ambient linear flow with $\boldsymbol{\omega}\cdot\boldsymbol{E}=0$. It is worth noting that the tensorial form of the inertial stress in (3.19) is independent of λ, the dependence on the latter only appearing in the multiplicative pre-factor. In § 5, we will use (3.19) specifically for the case of simple shear flow in order to examine the behaviour of the normal stress differences as a function of Re and Ca. The invariance of the tensorial form implies that the ratio of the $O(\phi Re)$ contributions to the two normal stress differences is independent of λ. Taking the limit $\lambda\to\infty$, and omitting the $O(\phi Ca)$ terms, one obtains the following expression for the excess stress, to $O(\phi Re)$, in a suspension of rigid particles:

$$\Sigma_{ij}^{(d)} = 5\phi E_{ij} - (\phi Re)\frac{4}{3}(\Omega_{ik}E_{jk}+\Omega_{jk}E_{ik}) + O(\phi^2, \phi Re^{3/2}), \tag{3.20}$$

a result identical to that obtained by Lin *et al.* (1970) and Stone *et al.* (2000). In (3.20), the first term, of course, gives rise to the Einstein viscosity correction in the Stokesian limit, while the second term is indicative of a non-Newtonian rheology in the presence of inertia. The non-Newtonian rheology for an inertial suspension, at $O(\phi Re)$, arises solely due to the anisotropy of the disturbance velocity field at $Re=0$.

4. Bulk stress in a dilute emulsion: a general linear flow

In this section, we derive an expression for the bulk stress in a dilute emulsion subject to a general linear flow and thereby extend the results of § 3 to the case of (nearly) linear flows with vortex stretching. For these flows, it is seen from the vorticity equation (see (2.23) in § 2) that one needs, in addition to the linear part, a term of $O(Re)$ with a cubic dependence on $\boldsymbol{r}$, or a time dependence at the same order, in order to balance the vortex stretching. Now, the inertial contributions to the bulk stress, at $O(\phi Re)$, arise from the fluid motion in a region around the drop of the order of its own size. This may be seen, for instance, from the convergence of the acceleration and Reynolds stress integrals evaluated in § 3 using the leading-order Stokes velocity fields. The corrections to these inertial stresses arising from $O(Re)$ unsteady/nonlinear terms in the ambient flow will therefore only be $O(Re^2)$, and may be neglected in the present calculation. This is, of course, because the unsteady or nonlinear terms in the ambient flow become important only at large distances from the drop of the order of an inertial screening length, and the linear term remains dominant at smaller length scales. Thus, from (3.8), one may write the following expression for the excess stress due to the drop phase in an arbitrary linear flow:

$$\Sigma_{ij}^{(d)} = \frac{3\phi}{4\pi}\int_{A_d^+}\left[\frac{1}{2}\left(\sigma_{ik}r_j + \sigma_{jk}r_i\right)n_k - \left(u_i n_j + u_j n_i\right)\right]\mathrm{d}A \\ + \phi Re\left[\frac{1}{5}\Gamma_{ik}\Gamma_{jk} - \frac{1}{5}(E_{jk}\Gamma_{ik} + E_{ik}\Gamma_{jk}) - \frac{(43\lambda^2 + 36\lambda + 8)}{35(\lambda+1)^2}E_{ik}E_{kj}\right]. \quad (4.1)$$

As seen in § 3, the most difficult part of the stress calculation is the determination of the $O(Re)$ correction to the stresslet integral in (4.1). Below, this is, therefore, done using an alternate approach based on the reciprocal theorem, so one only needs to know the velocity and pressure fields for appropriately chosen Stokes flow problems.

Before delving into the details of the stresslet calculation for a drop, it helps to first summarize the main difference between this analysis and a similar one for a rigid particle. The calculation of the stresslet, and hence the bulk stress, in dilute suspensions of rigid particles has been done before via a reciprocal theorem approach. As already mentioned, Stone *et al.* (2009) have adopted this approach for the case of an inertial suspension, while the same approach has, in fact, been used earlier in determining the stress in an inertialess suspension of charged spherical particles (Sherwood 1980). The test problem in the reciprocal theorem formulation for the above cases involves the (instantaneous) velocity and pressure fields due to a spherical particle deforming with a specified velocity at its boundary; the latter corresponds to that of an ambient extensional flow. Since the reciprocal theorem formulation relates to a pair of velocity and stress fields via surface and volume integrals, its success depends on the knowledge of the boundary conditions in both problems involved. This allows either the calculation of a particular surface integral or its reduction to the unknown rheological quantity desired—the stresslet in our case; see (4.2). This reduction is, however, not immediate when calculating the bulk stress in an inertial suspension. In the absence of inertia, a rigid spherical particle in a linear flow spins with exactly half the local ambient vorticity. At finite Re, the rate of spin in a general linear flow differs from $(1/2)\,\boldsymbol{\omega}$, a difference related to vortex stretching in the ambient flow. For small Re, this deviation is $O(Re)$, and applying the boundary condition requires knowledge of this angular velocity correction. Its determination requires, in principle, a knowledge of either the $O(Re)$ disturbance velocity and pressure fields or a separate reciprocal theorem identity for calculation of the angular velocity correction. However, a solid-body rotation, at any order in Re, corresponds to an antisymmetric velocity field, and therefore does not enter the calculation for the stresslet, the symmetric first moment of the surface force density.

Thus, knowing the surface velocity boundary condition within a solid-body rotation is sufficient, and the aforementioned test problem with the reciprocal theorem identity allows the calculation of the $O(Re)$ correction to the rigid particle stresslet in the same manner as the corresponding identity in the inertialess limit.

The above simplification, however, does not follow for the case of a viscous drop simply because the velocity on the surface of the drop is more complicated than a solid-body rotation. Thus, a reciprocal theorem formulation analogous to the one above, although a necessary first step in the stresslet calculation, still has a surface integral involving an unknown interfacial velocity. Note that the interfacial velocity at $Re = 0$ is, of course, known from the solution of the corresponding Stokes problem. However, one needs to know the interfacial velocity to $O(Re)$ in order to determine the inertial correction to the stresslet. In the following analysis, this unknown velocity field is determined via a second novel reciprocal theorem formulation. The test problem in this formulation involves a spherical drop, rather than a deforming particle, and wherein the flow in either phase is driven by a specified discontinuity in the tangential stress at the interface. We write down the reciprocal theorem identities both within and outside the drop, and a combination of the two, together with the stress discontinuity in the test problem, allows the determination of the integral, involving the unknown interfacial velocity, in the original reciprocal theorem formulation.

We now begin with the statement of the reciprocal theorem at finite Re (see Kim & Karrila 1991; Subramanian & Koch 2005):

$$\int_{A_d} \boldsymbol{n}\cdot\boldsymbol{\sigma}'\cdot\tilde{\boldsymbol{u}}\,\mathrm{d}S + Re\int_{V_f} \boldsymbol{f}'\cdot\tilde{\boldsymbol{u}}\,\mathrm{d}V = \int_{A_d} \boldsymbol{n}\cdot\tilde{\boldsymbol{\sigma}}\cdot\boldsymbol{u}'\mathrm{d}S. \tag{4.2}$$

Here, $(\boldsymbol{u}', \boldsymbol{\sigma}', \boldsymbol{f}')$ denote the disturbance velocity, stress and inertial forcing fields for the problem of interest, in our case, a neutrally buoyant drop suspended in an ambient linear flow ($\boldsymbol{u}^\infty = \boldsymbol{\Gamma}\cdot\boldsymbol{r}$) at small but finite Re; while $(\tilde{\boldsymbol{u}}, \tilde{\boldsymbol{\sigma}})$ correspond to an appropriate Stokes flow problem (the 'test' problem referred to above). Furthermore, A_d denotes the surface of the (spherical) drop and V_f denotes the exterior fluid domain. The above identity has been written anticipating that the corresponding surface integrals at infinity vanish and the volume integral in (4.2) remains absolutely convergent. This is indeed the case since all relevant disturbance velocity fields decay as $O(1/r^2)$ for large r and the corresponding stress fields as $O(1/r^3)$. Because the drop is neutrally buoyant, it translates with the velocity of the ambient linear flow at its centre to leading order. Thus, in a reference frame that translates with the neutrally buoyant drop, the inertial forcing associated with the perturbation $\boldsymbol{u}'$ is given by $\boldsymbol{f}' = (\partial\boldsymbol{u}'/\partial t) + \boldsymbol{u}'\cdot\nabla\boldsymbol{u}' + \boldsymbol{\Gamma}\cdot\boldsymbol{u}' + (\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot\nabla\boldsymbol{u}'$, where we have accordingly neglected an inertial term of the form $\boldsymbol{U}_d\cdot\nabla\boldsymbol{u}'$ corresponding to convection by a slip velocity $\boldsymbol{U}_d$. Even for a neutrally buoyant drop, however, there may arise a drift due to either an interaction with an adjacent boundary or in the case where $\boldsymbol{u}^\infty$ is a nonlinear function of $\boldsymbol{r}$ (Leal 1980). Any such drift velocity must be $O(Re)$ if it arises on account of inertia (as for a rigid particle), or $O(Ca)$ if it is related to drop deformation; the corresponding convection terms are $O(Re^2)$ and $O(ReCa)$, respectively, and may be neglected. Thus, the above expression for $\boldsymbol{f}'$ is consistent when determining the stresslet to $O(Re)$.

As explained earlier, the stresslet involves the symmetric first moment of the surface force density, $\boldsymbol{\sigma}\cdot\boldsymbol{n}$, and the test problem must therefore correspond to a particle, instantaneously spherical in shape, but deforming with a velocity field that is given by $-\tilde{\boldsymbol{E}}\cdot\boldsymbol{n}$, $\tilde{\boldsymbol{E}}$ being a symmetric traceless tensor. The disturbance velocity field in the test problem is of the form $\tilde{\boldsymbol{u}} = \mathscr{U}^E(\boldsymbol{r}) : \tilde{\boldsymbol{E}}$ with

$$\mathscr{U}^E(\boldsymbol{r}) = -\frac{\boldsymbol{Ir}}{r^5} + \frac{5}{2}\left(\frac{1}{r^2} - 1\right)\frac{\boldsymbol{rrr}}{r^5}, \tag{4.3}$$

and the corresponding surface force density given by $\tilde{\boldsymbol{\sigma}} \cdot \boldsymbol{n}|_{r=1} = -3\tilde{\boldsymbol{E}} \cdot \boldsymbol{n}$. Using (4.3) and the above expression for $\tilde{\boldsymbol{\sigma}} \cdot \boldsymbol{n}$ in (4.2) and noting that the resulting identity only holds for a symmetric traceless (but otherwise arbitrary) $\tilde{\boldsymbol{E}}$, one obtains

$$\begin{aligned}\frac{1}{2}\int_{A_d}\left[\sigma'_{ik}n_k r_j + \sigma'_{jk}n_k r_i - \frac{2}{3}\,\delta_{ij}(\sigma'_{kl}r_k n_l)\right]\mathrm{d}S \\ = \frac{Re}{2}\int_{V_f}\left[f'_l\mathscr{U}^E_{lij} + f'_l\mathscr{U}^E_{lji} - \frac{2}{3}\,\delta_{ij}\left(f'_l\mathscr{U}^E_{lkk}\right)\right]\mathrm{d}V - \frac{3}{2}\int_{A_d}(u'_i n_j + u'_j n_i)\,\mathrm{d}S.\end{aligned} \tag{4.4}$$

Using $\boldsymbol{\sigma}' = \boldsymbol{\sigma} - \boldsymbol{\sigma}^\infty$ and applying the divergence theorem to convert the surface integral involving the ambient stress to one over the volume V_d of the drop, (4.4) becomes

$$\begin{aligned}\frac{1}{2}\int_{A_d}\left[\sigma_{ik}n_k r_j + \sigma_{jk}n_k r_i - \frac{2}{3}\,\delta_{ij}(\sigma_{kl}r_k n_l)\right]\mathrm{d}S \\ = \frac{1}{2}\int_{V_d}\left[\frac{\partial\sigma^\infty_{ik}}{\partial r_k}r_j + \frac{\partial\sigma^\infty_{jk}}{\partial r_k}r_i - \frac{2}{3}\,\delta_{ij}\left(\frac{\partial\sigma^\infty_{kl}}{\partial r_k}r_l\right)\right]\mathrm{d}V + \frac{Re}{2}\int_{V_f}\left[f'_l\mathscr{U}^E_{lij} + f'_l\mathscr{U}^E_{lji}\right. \\ \left. - \frac{2}{3}\,\delta_{ij}(f'_l\mathscr{U}^E_{lkk})\right]\mathrm{d}V + \int_{V_d}\left[\sigma^\infty_{ij} - \frac{1}{3}\,\delta_{ij}\,\sigma^\infty_{kk}\right]\mathrm{d}V - \frac{3}{2}\int_{A_d}(u'_i n_j + u'_j n_i)\,\mathrm{d}S.\end{aligned} \tag{4.5}$$

For a rigid particle, the integral on the left-hand side in (4.5) is the stresslet $\boldsymbol{S}^P$. Using $\boldsymbol{\sigma}^\infty - \frac{1}{3}\boldsymbol{I}(\boldsymbol{\sigma}^\infty : \boldsymbol{I}) = 2\boldsymbol{E}$ in the third volume integral on the right-hand side, and that $\boldsymbol{u}' = \boldsymbol{\Omega}_p \wedge \boldsymbol{n} - \boldsymbol{\Gamma} \cdot \boldsymbol{n}$ at $r = 1$, one finally obtains the following simplified expression for the stresslet:

$$\begin{aligned}S^P_{ij} = \frac{1}{2}\int_{V_d}\left[\frac{\partial\sigma^\infty_{ik}}{\partial r_k}r_j + \frac{\partial\sigma^\infty_{jk}}{\partial r_k}r_i - \frac{2}{3}\,\delta_{ij}\left(\frac{\partial\sigma^\infty_{kl}}{\partial r_k}r_l\right)\right]\mathrm{d}V + 5\int_{V_d}E_{ij}\,\mathrm{d}V \\ + \frac{Re}{2}\int_{V_f}\left[f'_l\mathscr{U}^E_{lij} + f'_l\mathscr{U}^E_{lji} - \frac{2}{3}\,\delta_{ij}\left(f'_l\mathscr{U}^E_{lkk}\right)\right]\mathrm{d}V,\end{aligned} \tag{4.6}$$

where we have used $\int_{A_d}(u_i n_j + u_j n_i)\mathrm{d}S = 2\int_{V_d} e_{ij}\mathrm{d}V$, here $\boldsymbol{e}$ is the rate of strain corresponding to the velocity field $\boldsymbol{u}$. The above expression for the stresslet and the preceding arguments with regard to the validity of the formulation for an arbitrary ambient linear flow appear in Stone *et al.* (2009), who derive an expression for the bulk stress to $O(\phi Re)$ in a suspension of rigid particles subject to a general linear flow. We, again, note that the essential simplification that allows the determination of the stresslet for a rigid particle using (4.6) results from knowing $\boldsymbol{u}'$ on the particle surface in (4.5) to within a solid-body rotation field. This is not the case for a drop, however. From (4.5), we see that for a drop, a calculation of the stresslet to $O(Re)$ requires a knowledge of the velocity $\boldsymbol{u}'$ to the same order. Although $\boldsymbol{u}' \cdot \boldsymbol{n} = -\boldsymbol{u}^\infty \cdot \boldsymbol{n}$, a kinematic requirement for a steady spherical interface, the $O(Re)$ correction to the tangential component of $\boldsymbol{u}'$ is not known *a priori* and appears to necessitate a calculation of the $O(Re)$ velocity field as in §2. However, as will be seen, the tangential component, to $O(Re)$, may be obtained using a second reciprocal theorem formulation now applied both to the interior and exterior fluid domains and that again requires only a knowledge of Stokes velocity and pressure fields. Before doing this, we discuss the reciprocal theorem formulation, (4.5), in a little more detail, and finally write it in a form appropriate to an ambient (nearly) linear flow; see (4.9).

To begin with, we observe that the stresslet for a drop, $\boldsymbol{S}^D$, as defined in (4.1) differs from that for a solid particle. Including this difference in (4.5), one obtains the following relation for the stresslet $\boldsymbol{S}^D$:

$$S^D_{ij} = \frac{1}{2}\int_{V_d}\left[\frac{\partial\sigma^\infty_{ik}}{\partial r_k}r_j + \frac{\partial\sigma^\infty_{jk}}{\partial r_k}r_i - \frac{2}{3}\delta_{ij}\left(\frac{\partial\sigma^\infty_{kl}}{\partial r_k}r_l\right)\right]\mathrm{d}V + 2\int_{V_d}E_{ij}\,\mathrm{d}V \\ + \frac{Re}{2}\int_{V_f}\left[f'_l\mathscr{U}^E_{lij} + f'_l\mathscr{U}^E_{lji} - \frac{2}{3}\delta_{ij}\left(f'_l\mathscr{U}^E_{lkk}\right)\right]\mathrm{d}V - \frac{5}{2}\int_{A_d}(u'_in_j + u'_jn_i)\,\mathrm{d}S. \quad (4.7)$$

Restricting consideration to linear flows without vortex stretching for the moment, we note that the velocity gradient tensor $\boldsymbol{\Gamma}$ is constant and the inertial acceleration is therefore a linear function of $\boldsymbol{r}$. Thus, $\nabla\cdot\boldsymbol{\sigma}^\infty = Re\boldsymbol{\Gamma}\cdot(\boldsymbol{\Gamma}\cdot\boldsymbol{r})$, and (4.7) simplifies to

$$S^D_{ij} = \frac{8\pi}{3}E_{ij} + \frac{2\pi}{15}Re\left[\Gamma_{ik}\Gamma_{kj} + \Gamma_{jk}\Gamma_{ki} - \frac{2}{3}\delta_{ij}(\Gamma_{lk}\Gamma_{kl})\right] \\ + \frac{Re}{2}\int_{V_f}\left[f'_l\mathscr{U}^E_{lij} + f'_l\mathscr{U}^E_{lji} - \frac{2}{3}\delta_{ij}\left(f'_l\mathscr{U}^E_{lkk}\right)\right]\mathrm{d}V - \frac{5}{2}\int_{A_d}(u'_in_j + u'_jn_i)\,\mathrm{d}S. \quad (4.8)$$

The second term in (4.8) arises from the first moment of the $O(Re)$ acceleration in the ambient flow. Since $\boldsymbol{\omega}\cdot\boldsymbol{E} = 0$, such an acceleration may be balanced by the gradient of a quadratic pressure field. The latter was already encountered in § 2 in the expression for the $O(Re)$ correction to the exterior pressure field; see (2.20). Thus, the second term in (4.8) corresponds to the contribution, $(2\pi/15)(\Gamma_{ik}\Gamma_{kj} + \Gamma_{jk}\Gamma_{ki})$ in (3.17). For the case of simple shear flow, the pressure field is a constant and this term is identically zero.

When considering an ambient flow of the form $\boldsymbol{u}^\infty = \boldsymbol{\Gamma}\cdot\boldsymbol{r}$ for an arbitrary $\boldsymbol{\Gamma}$, as argued above, $\boldsymbol{\Gamma}$ is only a constant to $O(1)$. The flow is no longer linear at $O(Re)$, and one expects an additional $O(Re)$ quadratic inhomogeneity in the velocity gradient tensor besides a possible time dependence in the general case. In this case, it becomes convenient to shift the coordinate origin from the moving drop to a suitable fixed point. The centre of mass of the drop in this new reference frame is given by $\boldsymbol{x}^d(t)$, and the ambient flow in the vicinity of the drop may be written in the form $\boldsymbol{u}^\infty(\boldsymbol{x}, t) = \boldsymbol{u}^\infty(\boldsymbol{x}_d(t), t) + \boldsymbol{\Gamma}(\boldsymbol{x}_d(t), t)\cdot\boldsymbol{r}$ with $\boldsymbol{r} = \boldsymbol{x} - \boldsymbol{x}_d(t)$. Thus, $\nabla\cdot\boldsymbol{\sigma}^\infty = Re(\mathrm{D}^\infty\boldsymbol{u}^\infty)/(\mathrm{D}t)$, where $\mathrm{D}^\infty/\mathrm{D}t$ is the material derivative associated with the ambient flow. Following Stone *et al.* (2009), we Taylor-expand the velocity field about $\boldsymbol{x}_d(t)$ and perform the required surface integrations. The resulting stresslet in an arbitrary (nearly) linear flow may then be written as

$$S^D_{ij} = \frac{8\pi}{3}\left[E_{ij} + \frac{1}{10}\nabla^2E_{ij}\right]_{\boldsymbol{x}_d(t)} + \frac{2\pi}{15}Re\left[2\frac{\mathrm{D}^\infty E_{ij}}{\mathrm{D}t} + \left(\Gamma_{ik}\Gamma_{kj} + \Gamma_{jk}\Gamma_{ki} - \frac{2}{3}\delta_{ij}(\Gamma_{lk}\Gamma_{kl})\right)\right]_{\boldsymbol{x}_d(t)} \\ + \frac{Re}{2}\int_{V_f}\left[f'_l\mathscr{U}^E_{lij} + f'_l\mathscr{U}^E_{lji} - \frac{2}{3}\delta_{ij}\left(f'_l\mathscr{U}^E_{lkk}\right)\right]\mathrm{d}V - \frac{5}{2}\int_{A_d}(u'_in_j + u'_jn_i)\,\mathrm{d}S, \quad (4.9)$$

where the term proportional to $\nabla^2\boldsymbol{E}$ accounts for the first effects of inhomogeneity in the rate of strain; any higher-order variations are asymptotically smaller. Rewriting

(4.9) in terms of the rate of strain and vorticity tensors, one obtains

$$S_{ij}^D = \frac{8\pi}{3}\left[E_{ij} + \frac{1}{10}\nabla^2 E_{ij}\right]_{x_d(t)} + \frac{4\pi}{15}Re\left[\frac{D^\infty E_{ij}}{Dt} + \left(E_{ik}E_{jk} - \frac{1}{3}\delta_{ij}(E_{kl}E_{kl})\right)\right.$$
$$\left. + \left(\Omega_{ik}\Omega_{jk} - \frac{1}{3}\delta_{ij}(\Omega_{kl}\Omega_{kl})\right)\right]_{x_d(t)} + \frac{Re}{2}\int_{V_f}\left[f_l'\mathcal{U}_{lij}^E + f_l'\mathcal{U}_{lji}^E - \frac{2}{3}\delta_{ij}\left(f_l'\mathcal{U}_{lkk}^E\right)\right]\mathrm{d}V$$
$$-\frac{5}{2}\int_{A_d}(u_i'n_j + u_j'n_i)\,\mathrm{d}S, \tag{4.10}$$

which, of course, reduces to (4.8) for the case where $\boldsymbol{E}$ and $\boldsymbol{\Omega}$ are independent of spatial position. To $O(Re)$, the volume integral involving $\boldsymbol{f}'$ in (4.9) may be evaluated using the Stokes velocity field. This is, of course, equivalent to a solution via a regular perturbation expansion. As argued in § 2, the non-uniformity in the leading-order Stokes approximation and the related singular character of the inertial correction only enters at $O(\phi Re^{3/2})$. These observations apply to a rigid particle too. However, unlike a rigid particle, the integral $\int_{A_d}(\boldsymbol{u}'\boldsymbol{n} + \boldsymbol{u}'\boldsymbol{n})\,\mathrm{d}S$ still remains to be determined for the case of a drop. We again emphasize that the normal component of $\boldsymbol{u}'$ is, in fact, known to all orders in Re from the condition of a steady (spherical) interface. Thus, in what follows, we really aim to determine $\int_{A_d}(\boldsymbol{u}_t'\boldsymbol{n} + \boldsymbol{u}_t'\boldsymbol{n})\,\mathrm{d}S$, where $\boldsymbol{u}_t' = \boldsymbol{u}'\cdot(\boldsymbol{I} - \boldsymbol{nn})$ denotes the tangential component of $\boldsymbol{u}'$.

In order to determine $\int_{A_d}(\boldsymbol{u}_t'\boldsymbol{n} + \boldsymbol{u}_t'\boldsymbol{n})\,\mathrm{d}S$, we now consider the test problem of a drop in an otherwise quiescent fluid where the flow is driven by a stress jump at the interface. This stress jump may be specified by a symmetric traceless tensor $\hat{\boldsymbol{B}}$; the corresponding jump in the stress vector is $\hat{\boldsymbol{B}}\cdot\boldsymbol{n}$. The condition of zero trace arises because it is only the jump in tangential stress that drives a flow. There will be a corresponding jump in normal stress also needed in the calculation below. However, any shape deformation that results from the anisotropic part of this normal stress need not be accounted for, provided the relevant capillary number (defined in this case as $|\hat{\boldsymbol{B}}|a/T$) is small; we will assume this to be the case. Thus, as in § 2, the drop is assumed to be spherical, and the test velocity and pressure fields are determined with this assumption. The reciprocal theorem formulation is now applied to both the interior and exterior domains, and the viscosity ratio in the test problem must be equal to that in the actual problem, viz. λ. Denoting the exterior Stokes fields in the test problem by $(\tilde{\boldsymbol{u}}^+, \tilde{\sigma}^+)$ and those in the interior by $(\tilde{\boldsymbol{u}}^-, \tilde{\sigma}^-)$, one has the following relations for finite Re:

$$\int_{A_d}\boldsymbol{n}\cdot\tilde{\boldsymbol{\sigma}}^+\cdot\boldsymbol{u}'\mathrm{d}S = \int_{A_d}\boldsymbol{n}\cdot\boldsymbol{\sigma}'\cdot\tilde{\boldsymbol{u}}^+\,\mathrm{d}S + Re\int_{V_f}\boldsymbol{f}^+\cdot\tilde{\boldsymbol{u}}^+\,vV, \tag{4.11}$$

$$\int_{A_d}\hat{\boldsymbol{n}}\cdot\tilde{\boldsymbol{\sigma}}^-\cdot\boldsymbol{u}'\mathrm{d}S = \int_{A_d}\hat{\boldsymbol{n}}\cdot\boldsymbol{\sigma}'\cdot\tilde{\boldsymbol{u}}^-\,\mathrm{d}S + Re\int_{V_f}\boldsymbol{f}^-\cdot\tilde{\boldsymbol{u}}^-\,\mathrm{d}V. \tag{4.12}$$

Here, $\hat{\boldsymbol{n}} = -\boldsymbol{n}$ denotes the unit normal pointing into the drop and $(\boldsymbol{u}', \boldsymbol{\sigma}', \boldsymbol{f}^{\pm})$ still corresponds to a neutrally buoyant drop in a general linear flow at finite Re as in the earlier reciprocal theorem formulation, where $\boldsymbol{f}^+$ and $\boldsymbol{f}^-$, respectively, denote the inertial forcing fields in the exterior and the interior; thus,

$$\boldsymbol{f}^+ = \frac{\partial\boldsymbol{u}'}{\partial t} + \boldsymbol{u}'\cdot\nabla\boldsymbol{u}' + \boldsymbol{\Gamma}\cdot\boldsymbol{u}' + (\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot\nabla\boldsymbol{u}', \tag{4.13}$$

$$\boldsymbol{f}^- = \frac{1}{\lambda}\left[\frac{\partial\hat{\boldsymbol{u}}'}{\partial t} + \hat{\boldsymbol{u}}'\cdot\nabla\hat{\boldsymbol{u}}' + \boldsymbol{\Gamma}\cdot\hat{\boldsymbol{u}}' + (\boldsymbol{\Gamma}\cdot\boldsymbol{r})\cdot\nabla\hat{\boldsymbol{u}}'\right]. \tag{4.14}$$

Adding (4.11) and λ times (4.12), one obtains

$$\int_{A_d} \boldsymbol{n}\cdot(\tilde{\boldsymbol{\sigma}}^{+}-\lambda\tilde{\boldsymbol{\sigma}}^{-})\cdot\boldsymbol{u}'\,\mathrm{d}S = Re\left[\int_{V_f}\boldsymbol{f}^{+}\cdot\tilde{\boldsymbol{u}}^{+}\mathrm{d}V+\lambda\int_{V_d}\boldsymbol{f}^{-}\cdot\tilde{\boldsymbol{u}}^{-}\mathrm{d}V\right] + (\lambda-1)\int_{A_d}\boldsymbol{n}\cdot\boldsymbol{\sigma}^{\infty}\cdot\tilde{\boldsymbol{u}}|_{r=1}\,\mathrm{d}S, \quad (4.15)$$

where we have used the continuity of the tangential components of $\boldsymbol{\sigma}$ at $r=1$. The test velocity at $r=1$, $\tilde{\boldsymbol{u}}|_{r=1}$, is purely tangential and, therefore, the discontinuity in the normal stresses, $\boldsymbol{\sigma}':\boldsymbol{nn}$, does not enter the problem. Since the reciprocal theorem formulation is in terms of the perturbation fields, the continuity in the total tangential stress $\boldsymbol{\sigma}$ does not imply continuity of the perturbative tangential components ($\boldsymbol{\sigma}'$) that appear in (4.11) and (4.12). The difference is precisely the discontinuity in the ambient stress which appears on the right-hand side in (4.15). In the surface integral on the left-hand side of (4.15), we write $\boldsymbol{u}'$ as the sum of its normal and tangential components; that is, $\boldsymbol{u}'=\boldsymbol{u}'_t-(\boldsymbol{u}^{\infty}\cdot\boldsymbol{n})\boldsymbol{n}$. This has the effect of splitting the stress jump vector in the test problem into its tangential and normal components. The former has been specified; $\boldsymbol{n}\cdot(\tilde{\boldsymbol{\sigma}}^{+}-\lambda\tilde{\boldsymbol{\sigma}}^{-})\cdot(\boldsymbol{I}-\boldsymbol{nn})=\hat{\boldsymbol{B}}\cdot\boldsymbol{n}$. The latter must be determined as a part of the solution. Thus, (4.15) takes the form

$$\hat{\boldsymbol{B}}:\int_{A_d}\boldsymbol{n}\boldsymbol{u}'_t\,\mathrm{d}S-\int_{A_d}\{\boldsymbol{n}\cdot(\tilde{\boldsymbol{\sigma}}^{+}-\lambda\tilde{\boldsymbol{\sigma}}^{-})\cdot\boldsymbol{n}\}\mathsf{E}:\boldsymbol{nn}\,\mathrm{d}S = Re\left[\int_{V_f}\boldsymbol{f}^{+}\cdot\tilde{\boldsymbol{u}}^{+}\mathrm{d}V+\lambda\int_{V_d}\boldsymbol{f}^{-}\cdot\tilde{\boldsymbol{u}}^{-}\mathrm{d}V\right]+(\lambda-1)\int_{A_d}\boldsymbol{n}\cdot\boldsymbol{\sigma}^{\infty}\cdot\tilde{\boldsymbol{u}}|_{r=1}\,\mathrm{d}S. \quad (4.16)$$

Furthermore, the linearity of the Stokes flow problems allows one to write $\tilde{\boldsymbol{u}}^{\pm}=\boldsymbol{\mathscr{U}}^{B\pm}(\boldsymbol{r}):\hat{\boldsymbol{B}}$, $\tilde{\boldsymbol{u}}|_{r=1}=\boldsymbol{\mathscr{U}}^{B}(\boldsymbol{n}):\hat{\boldsymbol{B}}$, and the normal component of the stress jump, $\boldsymbol{n}\cdot\Delta\tilde{\boldsymbol{\sigma}}\cdot\boldsymbol{n}$, must evidently be of the form $\tilde{\boldsymbol{\Sigma}}:\hat{\boldsymbol{B}}$, where $\tilde{\boldsymbol{\Sigma}}$ may, without loss of generality, be taken as symmetric and traceless. Since the resulting equality holds for an arbitrary $\hat{\boldsymbol{B}}$, we finally have

$$\begin{aligned}\int_{A_d}(u'_{ti}n_j+u'_{tj}n_i)\,\mathrm{d}S = &\int_{A_d}\tilde{\Sigma}_{ij}(\mathsf{E}:\boldsymbol{nn})\,\mathrm{d}S \\ &+2(\lambda-1)\int_{A_d}\left[E_{lm}\mathscr{U}^{B}_{lij}+E_{lm}\mathscr{U}^{B}_{lji}-\frac{2}{3}\delta_{ij}\left(E_{lm}\mathscr{U}^{B}_{lkk}\right)\right]n_m|_{r=1}\,\mathrm{d}S \\ &Re\left[\int_{V_f}\left[f^{+}_l\mathscr{U}^{B+}_{lij}+f^{+}_l\mathscr{U}^{B+}_{lji}-\frac{2}{3}\delta_{ij}\left(f^{+}_l\mathscr{U}^{B+}_{lkk}\right)\right]\mathrm{d}V\right. \\ &\left.+\lambda\int_{V_d}\left[f^{-}_l\mathscr{U}^{B-}_{lij}+f^{-}_l\mathscr{U}^{B-}_{lji}-\frac{2}{3}\delta_{ij}\left(f^{-}_l\mathscr{U}^{B-}_{lkk}\right)\right]\mathrm{d}V\right], \end{aligned} \quad (4.17)$$

for the surface integral of interest. As shown below, the first two terms on the right-hand side contribute to the Taylor viscosity, while the remaining two contribute to the $O(Re)$ corrections. To the order of approximation required, all quantities on the right-hand side may be evaluated using Stokes flow velocity and pressure fields, and the result may then be used in (4.9) to obtain the stresslet in an ambient linear flow. Doing so, one obtains the following general expression for the stresslet in a dilute emulsion:

$$
\begin{aligned}
S_{ij}^{D} =& \frac{8\pi}{3}\left[E_{ij} + \frac{1}{10}\nabla^2 E_{ij}\right]_{\boldsymbol{x}_d(t)} - \frac{5}{2}\int_{A_d} \tilde{\Sigma}_{ij}(\boldsymbol{E}:\boldsymbol{nn})\,\mathrm{d}S \\
&+ \frac{4\pi}{15}Re\left[\frac{\mathrm{D}^{\infty}E_{ij}}{\mathrm{D}t} + \left(E_{ik}E_{jk} - \frac{1}{3}\,\delta_{ij}(E_{kl}E_{kl})\right) + \left(\Omega_{ik}\Omega_{jk} - \frac{1}{3}\,\delta_{ij}(\Omega_{kl}\Omega_{kl})\right)\right]_{\boldsymbol{x}_d(t)} \\
&- 5(\lambda-1)\int_{A_d}\left[E_{lm}\mathscr{U}_{lij}^{B} + E_{lm}\mathscr{U}_{lji}^{B} - \frac{2}{3}\,\delta_{ij}\left(E_{lm}\mathscr{U}_{lkk}^{B}\right)\right] n_m|_{r=1}\mathrm{d}S \\
&+ Re\left[\frac{1}{2}\int_{V_f}\left[f_l'\mathscr{U}_{lij}^{E} + f_l'\mathscr{U}_{lji}^{E} - \frac{2}{3}\,\delta_{ij}\left(f_l'\mathscr{U}_{lkk}^{E}\right)\right]\mathrm{d}V\right. \\
&-\frac{5}{2}\int_{V_f}\left[f_l^{+}\mathscr{U}_{lij}^{B+} + f_l^{+}\mathscr{U}_{lji}^{B+} - \frac{2}{3}\,\delta_{ij}\left(f_l^{+}\mathscr{U}_{lkk}^{B+}\right)\right]\mathrm{d}V \\
&\left.-\frac{5}{2}\lambda\int_{V_d}\left[f_l^{-}\mathscr{U}_{lij}^{B-} + f_l^{-}\mathscr{U}_{lji}^{B-} - \frac{2}{3}\,\delta_{ij}\left(f_l^{-}\mathscr{U}_{lkk}^{B-}\right)\right]\mathrm{d}V\right]. \qquad (4.18)
\end{aligned}
$$

The solution to the test problem is straightforward, and we include a brief description of the same in Appendix C. One finds

$$\mathscr{U}_{lij}^{B+} = \frac{1}{(\lambda+1)}\left[\left(-\frac{3}{10r^5} + \frac{1}{2r^7}\,r_i r_j r_l\right) - \frac{\delta_{lj} r_i}{5r^5}\right], \qquad (4.19)$$

$$\mathscr{U}_{lij}^{B-} = \frac{1}{(\lambda+1)}\left[\left(\frac{3}{10} - \frac{r^2}{2}\right)\delta_{lj} r_i + \frac{1}{5}\,r_i r_j r_l\right], \qquad (4.20)$$

$$\tilde{\Sigma}_{ij} = -\frac{(9\lambda+6)}{(\lambda+1)} n_i n_j, \qquad (4.21)$$

$$\mathscr{U}_{lij}^{B} = \frac{1}{5(\lambda+1)}[n_i n_j n_l - \delta_{lj} n_i], \qquad (4.22)$$

now to be used in (4.18). First, evaluating the $O(1)$ contributions in (4.18), one finds

$$\int_{A_d} \tilde{\Sigma}_{ij}(\boldsymbol{E}:\boldsymbol{nn})\,\mathrm{d}S = -\frac{4\pi}{25}\frac{(3\lambda+2)}{(\lambda+1)}E_{ij}, \qquad (4.23)$$

$$2(\lambda-1)\int_{A_d}\left[E_{lm}\mathscr{U}_{lij}^{B} + E_{lm}\mathscr{U}_{lji}^{B} - \frac{2}{3}\,\delta_{ij}\left(E_{lm}\mathscr{U}_{lkk}^{B}\right)\right] n_m|_{r=1}\,\mathrm{d}S = -\frac{8\pi(\lambda-1)}{25(\lambda+1)}E_{ij}. \qquad (4.24)$$

Using (4.23) and (4.24), (4.18) takes the form

$$
\begin{aligned}
S_{ij}^{D} =& \frac{8\pi}{3}\left[\frac{(5\lambda+2)}{2(\lambda+1)}E_{ij} + \frac{1}{10}\nabla^2 E_{ij}\right]_{\boldsymbol{x}_d(t)} \\
&+ \frac{4\pi}{15}Re\left[\frac{\mathrm{D}^{\infty}E_{ij}}{\mathrm{D}t} + \left(E_{ik}E_{jk} - \frac{1}{3}\,\delta_{ij}(E_{kl}E_{kl})\right) + \left(\Omega_{ik}\Omega_{jk} - \frac{1}{3}\,\delta_{ij}(\Omega_{kl}\Omega_{kl})\right)\right]_{\boldsymbol{x}_d(t)} \\
&+ Re\left[\frac{1}{2}\int_{V_f}\left[f_l'\mathscr{U}_{lij}^{E} + f_l'\mathscr{U}_{lji}^{E} - \frac{2}{3}\,\delta_{ij}\left(f_l'\mathscr{U}_{lkk}^{E}\right)\right]\mathrm{d}V\right. \\
&-\frac{5}{2}\int_{V_f}\left[f_l^{+}\mathscr{U}_{lij}^{B+} + f_l^{+}\mathscr{U}_{lji}^{B+} - \frac{2}{3}\,\delta_{ij}\left(f_l^{+}\mathscr{U}_{lkk}^{B+}\right)\right]\mathrm{d}V \\
&\left.-\frac{5}{2}\lambda\int_{V_d}\left[f_l^{-}\mathscr{U}_{lij}^{B-} + f_l^{-}\mathscr{U}_{lji}^{B-} - \frac{2}{3}\,\delta_{ij}\left(f_l^{-}\mathscr{U}_{lkk}^{B-}\right)\right]\mathrm{d}V\right], \qquad (4.25)
\end{aligned}
$$

where the first term includes both the Newtonian response for a linear flow that may be interpreted in terms of a Taylor viscosity and a Faxen-like correction for an inhomogeneous rate of strain field. Using (4.3), (4.19) and (4.20), and the definitions of $\boldsymbol{f}^{\pm}$ in (4.13) and (4.14), we now calculate the three volume integrals in (4.25). The contributions to the stresslet arising from each of the terms in $\boldsymbol{f}^{\pm}$ are as follows:

$$\left.\begin{aligned} &\int_{V_f} \frac{\partial u'_l}{\partial t}\left[\mathscr{U}^E_{lij} + \mathscr{U}^E_{lji} - \frac{2}{3}\,\delta_{ij}\left(\mathscr{U}^E_{lkk}\right)\right]\mathrm{d}V = \frac{8\pi(9\lambda+4)}{15(\lambda+1)}\frac{\partial E_{ij}}{\partial t},\\ &\int_{V_f} \varGamma_{lm}u'_m\left[\mathscr{U}^E_{lij} + \mathscr{U}^E_{lji} - \frac{2}{3}\,\delta_{ij}\left(\mathscr{U}^E_{lkk}\right)\right]\mathrm{d}V = \frac{4\pi(7\lambda+2)}{45(\lambda+1)}(\varOmega_{ik}E_{jk}+\varOmega_{jk}E_{ik})\\ &\quad+\frac{8\pi(43\lambda+18)}{105(\lambda+1)}E_{ik}E_{jk} - \frac{8\pi(43\lambda+18)}{315(\lambda+1)}(E_{kl}E_{kl})\delta_{ij}, \end{aligned}\right\} \tag{4.26}$$

$$\begin{aligned} &\int_{V_f} \varGamma_{nm}r_m\frac{\partial u'_m}{\partial r_n}\left[\mathscr{U}^E_{lij} + \mathscr{U}^E_{lji} - \frac{2}{3}\,\delta_{ij}\left(\mathscr{U}^E_{lkk}\right)\right]\mathrm{d}V = \frac{4\pi(47\lambda+22)}{45(\lambda+1)}(\varOmega_{ik}E_{jk}+\varOmega_{jk}E_{ik})\\ &\quad+\frac{8\pi(7\lambda+6)}{105(\lambda+1)}E_{ik}E_{jk} - \frac{8\pi(7\lambda+6)}{315(\lambda+1)}(E_{kl}E_{kl})\delta_{ij}, \end{aligned} \tag{4.27}$$

$$\begin{aligned} &\int_{V_f} u_n\frac{\partial u'_m}{\partial r_n}\left[\mathscr{U}^E_{lij} + \mathscr{U}^E_{lji} - \frac{2}{3}\,\delta_{ij}\left(\mathscr{U}^E_{lkk}\right)\right]\mathrm{d}V = \frac{8\pi(7\lambda^2+6)}{105(\lambda+1)}E_{ik}E_{jk}\\ &\quad-\frac{8\pi(7\lambda+6)}{315(\lambda+1)}(E_{kl}E_{kl})\delta_{ij}, \end{aligned} \tag{4.28}$$

$$\left.\begin{aligned} &\int_{V_f} \frac{\partial u'_l}{\partial t}\left[\mathscr{U}^{B+}_{lij} + \mathscr{U}^{B+}_{lji} - \frac{2}{3}\,\delta_{ij}\left(\mathscr{U}^{B+}_{lkk}\right)\right]\mathrm{d}V = \frac{8\pi(5\lambda+2)}{75(\lambda+1)^2}\frac{\partial E_{ij}}{\partial t},\\ &\int_{V_f} \varGamma_{lm}u'_m\left[\mathscr{U}^{B+}_{lij} + \mathscr{U}^{B+}_{lji} - \frac{2}{3}\,\delta_{ij}\left(\mathscr{U}^{B+}_{lkk}\right)\right]\mathrm{d}V\\ &\quad= \frac{(5\lambda+2)}{(\lambda+1)^2}\left[\frac{4\pi}{225}(\varOmega_{ik}E_{jk}+\varOmega_{jk}E_{ik}) + \frac{8\pi}{105}E_{ik}E_{jk} - \frac{8\pi}{315}(E_{kl}E_{kl})\delta_{ij}\right], \end{aligned}\right\} \tag{4.29}$$

$$\begin{aligned} &\int_{V_f} \varGamma_{nm}r_m\frac{\partial u'_l}{\partial r_n}\left[\mathscr{U}^{B+}_{lij} + \mathscr{U}^{B+}_{lji} - \frac{2}{3}\,\delta_{ij}\left(\mathscr{U}^{B+}_{lkk}\right)\right]\mathrm{d}V\\ &\quad= \frac{(5\lambda+2)}{(\lambda+1)^2}\left[\frac{4\pi}{45}(\varOmega_{ik}E_{jk}+\varOmega_{jk}E_{ik}) - \frac{8\pi}{525}E_{ik}E_{jk} + \frac{8\pi}{1575}(E_{kl}E_{kl})\delta_{ij}\right], \end{aligned} \tag{4.30}$$

$$\begin{aligned} &\int_{V_f} u'_n\frac{\partial u'_l}{\partial r_n}\left[\mathscr{U}^{B+}_{lij} + \mathscr{U}^{B+}_{lji} - \frac{2}{3}\,\delta_{ij}\left(\mathscr{U}^{B+}_{lkk}\right)\right]\mathrm{d}V\\ &\quad= -\frac{8\pi(5\lambda+2)}{225(\lambda+1)^2}E_{ik}E_{jk} - \frac{8\pi(5\lambda+2)}{1575(\lambda+1)^2}(E_{kl}E_{kl})\delta_{ij}, \end{aligned} \tag{4.31}$$

$$\left.\begin{aligned} &\int_{V_d} \frac{\partial \hat{u}'_l}{\partial t}\left[\mathscr{U}^{B-}_{lij} + \mathscr{U}^{B-}_{lji} - \frac{2}{3}\,\delta_{ij}\left(\mathscr{U}^{B-}_{lkk}\right)\right]\mathrm{d}V = -\frac{8\pi}{225(\lambda+1)^2}\frac{\partial E_{ij}}{\partial t},\\ &\int_{V_d} \varGamma_{lm}\hat{u}'_m\left[\mathscr{U}^{B-}_{lij} + \mathscr{U}^{B-}_{lji} - \frac{2}{3}\,\delta_{ij}\left(\mathscr{U}^{B-}_{lkk}\right)\right]\mathrm{d}V\\ &\quad= \frac{1}{(\lambda+1)^2}\left[-\frac{4\pi}{675}(\varOmega_{ik}E_{jk}+\varOmega_{jk}E_{ik}) - \frac{8\pi}{315}E_{ik}E_{jk} + \frac{8\pi}{945}(E_{kl}E_{kl})\delta_{ij}\right], \end{aligned}\right\} \tag{4.32}$$

$$\int_{V_d} \Gamma_{nm} r_m \frac{\partial \hat{u}_l}{\partial r_n}\left[\mathscr{U}^{B-}_{lij} + \mathscr{U}^{B-}_{lji} - \frac{2}{3}\delta_{ij}\left(\mathscr{U}^{B-}_{lkk}\right)\right]\mathrm{d}V$$
$$= \frac{1}{(\lambda+1)^2}\left[-\frac{4\pi}{135}(\Omega_{ik}E_{jk}+\Omega_{jk}E_{ik}) - \frac{8\pi}{175}E_{ik}E_{jk} + \frac{8\pi}{525}(E_{kl}E_{kl})\delta_{ij}\right], \quad (4.33)$$

$$\int_{V_d} \hat{u}'_n \frac{\partial \hat{u}_l}{\partial r_n}\left[\mathscr{U}^{B-}_{lij} + \mathscr{U}^{B-}_{lji} - \frac{2}{3}\delta_{ij}\left(\mathscr{U}^{B-}_{lkk}\right)\right]\mathrm{d}V$$
$$= \frac{1}{(\lambda+1)^2}\left[-\frac{4\pi}{675}(\Omega_{ik}E_{jk}+\Omega_{jk}E_{ik}) - \frac{8\pi}{315}E_{ik}E_{jk} + \frac{8\pi}{945}(E_{kl}E_{kl})\delta_{ij}\right]. \quad (4.34)$$

Using the above results in (4.18) in the lab reference frame (so $\partial \boldsymbol{E}/\partial t$ in the above expressions becomes the convected derivative $(\mathrm{D}^\infty E)/(\mathrm{D}t)$) one obtains the following expression for the stresslet in a general linear flow:

$$\begin{aligned} S^D_{ij} =& \frac{8\pi}{3}\left[\frac{(5\lambda+2)}{2(\lambda+1)}E_{ij} + \frac{1}{10}\nabla^2 E_{ij}\right]_{\boldsymbol{x}_d(t)} \\ &+ \frac{4\pi}{15}Re\left[\frac{\mathrm{D}^\infty E_{ij}}{\mathrm{D}t} + \left(E_{ik}E_{jk} - \frac{1}{3}\delta_{ij}(E_{kl}E_{kl})\right) + \left(\Omega_{ik}\Omega_{jk} - \frac{1}{3}\delta_{ij}(\Omega_{kl}\Omega_{kl})\right)\right]_{\boldsymbol{x}_d(t)} \\ &+ Re\left[\frac{4\pi(27\lambda^2+24\lambda+7)}{45(\lambda+1)^2}\frac{\mathrm{D}^\infty E_{ij}}{\mathrm{D}t} - \frac{16\pi(3\lambda^2+3\lambda+1)}{27(\lambda+1)^2}(\Omega_{ik}E_{jk}+\Omega_{jk}E_{ik}) \right. \\ &\left. + \frac{4\pi(43\lambda^2+36\lambda+8)}{105(\lambda+1)^2}\left(E_{ik}E_{jk} - \frac{1}{3}\delta_{ij}(E_{kl}E_{kl})\right)\right]_{\boldsymbol{x}_d(t)}. \end{aligned} \quad (4.35)$$

Furthermore, combining (4.35) with the expression (3.16) for the inertial stresses, derived earlier in § 3 and valid for a general linear flow, one obtains the corresponding expression for the excess disperse phase stress to $O(\phi Re)$:

$$\begin{aligned} \Sigma^{(d)}_{ij} = 2\phi&\left[\frac{(5\lambda+2)}{2(\lambda+1)}E_{ij} + \frac{1}{10}\nabla^2 E_{ij}\right]_{\boldsymbol{x}_d(t)} + (\phi Re)\frac{2(3\lambda^2+3\lambda+1)}{3(\lambda+1)^2} \\ &\times\left[\frac{\mathrm{D}^\infty E_{ij}}{\mathrm{D}t} - \frac{2}{3}(\Omega_{ik}E_{jk}+\Omega_{jk}E_{ik})\right] + O(\phi^2, \phi Re^{3/2}, \phi Ca). \end{aligned} \quad (4.36)$$

For the case where $\boldsymbol{E}$ and $\boldsymbol{\Omega}$ are constants, (4.36) reduces to the expression calculated in § 3 for linear flows without vortex stretching, thereby validating the correctness of the rather tedious perturbation analysis. Furthermore, for the case discussed in § 2, where the weak (cubic) nonlinearity or time dependence in the ambient flow is induced by the vortex stretching in the dominant linear part, both $\partial \boldsymbol{E}/\partial t$ and $\nabla^2 \boldsymbol{E}$ are $O(Re)$; the term $\boldsymbol{u}^\infty \cdot \nabla \boldsymbol{E}$ is $O(Re^2)$, and the convected derivative of the rate of strain in (4.36) may be replaced by the Eulerian derivative. However, this is merely one instance of a nonlinear ambient flow, and there exist other ambient flows in which the convection of the inhomogeneous rate of strain field remains of the same order as in the other terms. For instance, a turbulent flow field, on the scale of a sub-Kolmogorov drop, may be written in the (dimensional) form $\boldsymbol{u}^\infty = \dot{\gamma}_\kappa \boldsymbol{\Gamma}\cdot\boldsymbol{x} + O(\dot{\gamma}_\kappa/l_\kappa)(\boldsymbol{Q}:\boldsymbol{xx})$, here $\boldsymbol{Q}$ is a third-order tensor. The quadratic nonlinearity is again smaller on length scales of the drop size, but by a geometric factor of $O(a/l_\kappa)$ instead; thus, both $\boldsymbol{u}^\infty \cdot \nabla \boldsymbol{E}$ and terms of the form $\boldsymbol{E}\cdot\boldsymbol{E}$ are now $O(\dot{\gamma}_\kappa)^2$. For a general nonlinear ambient flow, the convected derivative has, therefore, to be retained in its full form. It is again worth emphasizing that the tensorial form of the bulk stress at $O(\phi Re)$ remains identical to that for an inertial suspension; the numerical pre-factor is now a function of λ.

The above expression for the stress is valid for an arbitrary flow that remains slow and slowly varying on the microscale. Thus, it is of interest to compare (4.36) with the retarded motion expansion known to model the rheology of polymer solutions and melts in the limit of small Deborah number (see Bird *et al.* 1987; Larson 1988). In fact, the rheology of an infinitely dilute inertialess emulsion is expected to conform to the general structure of the retarded motion expansion to all orders with Ca playing the role of the Deborah number. The singular nature of the inertial contribution at $O(Re^{3/2})$ ensures that the structure of a similar expansion for an inertial emulsion is much more complicated starting at this order. However, the effect of inertia, even at $O(Re)$, is non-trivial. The relevant comparison in this case would be with the retarded motion expansion truncated at quadratic order, that is, the constitutive relation for a second-order fluid given by

$$\boldsymbol{\Sigma}^{second-order} = 2\boldsymbol{E} - \Psi_1\left(\frac{D\boldsymbol{E}}{Dt} + \boldsymbol{\Omega}\cdot\boldsymbol{E} - \boldsymbol{E}\cdot\boldsymbol{\Omega}\right) + (2\Psi_1 + 4\Psi_2)\boldsymbol{E}\cdot\boldsymbol{E}, \tag{4.37}$$

where Ψ_1 and Ψ_2 are the normal stress coefficients. Note that the numerical coefficient multiplying the co-rotational terms ($\boldsymbol{\Omega}\cdot\boldsymbol{E} - \boldsymbol{E}\cdot\boldsymbol{\Omega}$) is the same as the convected derivative, and this is fixed by the requirement of frame invariance (otherwise known as the principle of material frame indifference); any rigid-body rotation only serves to rotate the principal axes of the stress-inducing microstructure (Larson 1988). Notwithstanding the non-locality inherent in the Faxen-like term, one may now compare (4.36) and (4.37). We note thereby that (4.36) is not frame invariant. Thus, as pointed out earlier in the context of turbulence (Lumley 1970) and inertial suspensions (see Ryskin 1980; Stone *et al.* 2000), the constitutive relation for a dilute inertial emulsion is another example where microscale inertia leads to a violation of the principle of material frame indifference. This also implies, as is also evident physically, that the constitutive equation for one linear flow may not be deduced from another by the mere use of arguments based on symmetry and invariance. Finally, we note that the validity of (4.36) for rheologically complex flows would allow one to examine flow scenarios that often arise in a microfluidic context, such as flow past obstacles, flow through wavy-walled tubes or over topography (the purpose of which might be to aid mixing) and flow through a contraction.

5. Emulsion rheology in simple shear and extensional flows

In this section, we focus on the rheology of an emulsion in two canonical flows—simple shear flow and extensional flow (in both cases, $\boldsymbol{\omega}\cdot\boldsymbol{E}=0$, and the results of § 3 may be used). For simple shear flow, $\boldsymbol{E}=(1/2)(\mathbf{1}_x\mathbf{1}_y + \mathbf{1}_y\mathbf{1}_x)$ and $\boldsymbol{\omega}=-\mathbf{1}_z$. Using (3.12), one obtains, to $O(\phi Ca)$, the following expressions for the two normal stress differences:

$$N_1^{(0)} = \Sigma_{xx} - \Sigma_{yy} = \phi Ca\,\frac{1}{40}\left(\frac{19\lambda+16}{\lambda+1}\right)^2, \tag{5.1}$$

$$N_2^{(0)} = \Sigma_{yy} - \Sigma_{zz} = -\phi Ca\,\frac{(551\lambda^3 + 1623\lambda^2 + 1926\lambda + 800)}{280(\lambda+1)^3}, \tag{5.2}$$

for a dilute emulsion in the absence of inertia. Thus, N_1 is positive and N_2 negative. The signs of the normal stress differences may be related to the deformation of the drop in the imposed shear flow. At $O(Ca)$, the originally spherical drop is stretched into an ellipsoid with its major axis oriented along the extensional axis of the simple shear. The drop deformation at this order is proportional to the rate of strain tensor, since the shape of a spherical drop remains unaffected by rotation. This then leads to a Newtonian response at $O(\phi)$. At $O(Ca^2)$, however, the vorticity in the ambient simple shear acts to rotate the ellipsoid towards the flow direction, and the resulting tensile component of the surface tension forces in this direction gives rise to a positive N_1 at $O(\phi Ca)$. On the other hand, the additional compressive stress in the gradient direction leads to a negative N_2 at the same order.

Using (3.19), and the normal stress differences corresponding to each of the tensorial terms therein, viz. $N_1^{\boldsymbol{E}\cdot\boldsymbol{E}}=0$, $N_2^{\boldsymbol{E}\cdot\boldsymbol{E}}=1/4$ and $N_1^{\boldsymbol{\Omega}\cdot\boldsymbol{E}-\boldsymbol{E}\cdot\boldsymbol{\Omega}}=1$, $N_2^{\boldsymbol{\Omega}\cdot\boldsymbol{E}-\boldsymbol{E}\cdot\boldsymbol{\Omega}}=-1/2$, one may now write down the following expressions for N_1 and N_2 in the presence of microscale inertia:

$$N_1 = N_1^{(0)} - \phi Re\frac{4(3\lambda^2+3\lambda+1)}{9(\lambda+1)^2} + O(\phi^2, \phi Ca^2, \phi Re^{3/2}), \tag{5.3}$$

$$N_2 = N_2^{(0)} + \phi Re\frac{2(3\lambda^2+3\lambda+1)}{9(\lambda+1)^2} + O(\phi^2, \phi Ca^2, \phi Re^{3/2}). \tag{5.4}$$

The contributions at $O(\phi Ca)$ and (ϕRe) are evidently of opposite signs. It is seen from (3.16) that the direct inertial contributions (i.e. the Reynolds and acceleration stresses) only contribute to an N_2 via the λ-dependent term proportional to $\boldsymbol{E}\cdot\boldsymbol{E}$. However, as already mentioned, this term is identically cancelled by a term of the same form in the stresslet contribution; see (3.17). Therefore, the $O(\phi Re)$ contributions to both N_1 (positive) and N_2 (negative) arise entirely from the stresslet integral. Furthermore, since the contributing term is proportional to $\boldsymbol{\Omega}\cdot\boldsymbol{E}-\boldsymbol{E}\cdot\boldsymbol{\Omega}$, the ratio of the $O(\phi Re)$ contributions to N_1 and N_2 is independent of λ, being equal to -2.

The underlying physics becomes clearer from examining the alternate expression, (3.5), for the bulk stress, rather than (3.3) used for the above calculation. In (3.5), the contribution to the normal stress differences from the Reynolds stresses arises because of the anisotropy of the Stokes velocity field fluctuations. Since the disturbance velocity field at $Re=0$ is only a function of $\boldsymbol{E}$, the Reynolds stress contribution must be proportional to $\boldsymbol{E}\cdot\boldsymbol{E}$. Thus, the reversed signs of the normal stress differences at $O(\phi Re)$ are related to the first two terms in (3.5), that is, the $O(Re)$ modification of the viscous and interfacial tension stresses acting on the drop surface; the latter arise because of the deformation of the drop at $O(ReCa)$ on account of inertial forces. Now, for $\lambda=1$, the viscous stress contribution is identically zero, and the reversal in the sign of the normal stress differences for this viscosity ratio is therefore a direct result of the dominance of the inertial $O(ReCa)$ drop deformation over the $O(Ca^2)$ deformation induced by viscous stresses. Indeed, it may be shown using the normal stress balance at $O(Re)$ that the drop deformation at $O(ReCa)$ acts to rotate it towards the velocity gradient direction. This is in contrast to the flow-aligning deformation at $O(Ca^2)$. The opposing roles of inertial and viscous forces with regard to drop deformation in simple shear, for a viscosity ratio of unity, have already been highlighted in the numerical simulations of Renardy & Cristini (2001) and Li & Sarkar (2005); the latter simulations are compared in some detail with the analytical predictions obtained here at the end of this section. Determining the relative magnitudes of the $O(Re)$ viscous and interfacial tension contributions for other viscosity ratios would entail a separate calculation, to $O(Re)$, of the individual terms in the expression (3.5) for the stresslet. Although this is not done here, it is worth mentioning the relative magnitudes of the analogous contributions to the shear viscosity for $Re=0$. Again, using (3.5), it is easily shown that the viscous and interfacial tension contributions to the Taylor viscosity are $(19\lambda+16)/10(\lambda+1)$ and $\{3(\lambda-1)\}/\{5(\lambda+1)\}$, respectively, and the ratio of the two contributions, $(19\lambda+16)/6(\lambda-1)$, remains of order unity for all viscosity ratios and diverges for $\lambda \to 1$. In fact, for drops of lower viscosity $(\lambda<1)$, the viscous contribution changes sign, but the interfacial tension contribution (because of drop deformation along the extensional axis) overwhelms this reduction and ensures that a dilute emulsion, to $O(\phi)$, is always more viscous than the suspending fluid. Thus, it does seem reasonable to conclude, even for small but finite Re, that it is the nature of the $O(ReCa)$ drop deformation and the resulting anisotropy of the surface tension forces that is primarily responsible for the reversal in rheological characteristics; this is expected to be so at least for a range of viscosity ratios around unity. Finally, we also note that the effective shear viscosity of the emulsion remains unchanged to the order (quadratic) considered in the analysis; thus, to $O(\phi Re)$ or $O(\phi Ca)$, the shear viscosity of a dilute emulsion is still given by the Taylor result (Taylor 1932).

From (5.3), one finds that N_1 changes sign at a critical Reynolds number given by

$$Re_c^{N1} = \frac{9(19\lambda+16)^2}{160(3\lambda^2+3\lambda+1)}Ca, \tag{5.5}$$

where N_1 is positive for $Re < Re_c^{N1}$ and negative for higher Re. Similarly, using (5.4), the critical Reynolds number for N_2 is found to be

$$Re_c^{N2} = \frac{9(551\lambda^3+1623\lambda^2+1926\lambda+800)}{560(\lambda+1)(3\lambda^2+3\lambda+1)}Ca, \tag{5.6}$$

the change in the sign in this case being from negative to positive with increasing Re. It is worth noting that provided $Ca \ll 1$, the predictions (5.5) and (5.6) for the critical Reynolds numbers are consistent with the assumption of weak inertia implicit in the perturbation analysis of § 2. While the formulation of the threshold conditions in terms of a critical Reynolds number is useful when comparing with simulation results (see below), it is important to note that the threshold condition for a reversal in the sign of either N_1 or N_2 is independent of the shear rate. Thus, from an experimental point of view, (5.5) and (5.6) are more conveniently written in terms of a critical Ohnesorge number (Oh_c), a dimensionless parameter that is only a function of the system properties, as a function of the viscosity ratio. The Ohnesorge number being defined as $Oh=(Ca/Re)^{1/2}$, one finds the following critical values corresponding to (5.5) and (5.6):

$$Oh_c^{N1} = \frac{4\sqrt{10}}{3}\frac{(3\lambda^2+3\lambda+1)^{1/2}}{(19\lambda+16)}, \tag{5.7}$$

$$Oh_c^{N2} = \frac{4}{3}\left[\frac{35(\lambda+1)(3\lambda^2+3\lambda+1)}{(551\lambda^3+1623\lambda^2+1926\lambda+800)}\right]^{1/2}. \tag{5.8}$$

The ratio of the two Ohnesorge numbers is given by

$$\frac{Oh_c^{N2}}{Oh_c^{N1}} = \left[\frac{7(\lambda+1)(19\lambda+16)}{2(29\lambda^2+61\lambda+50)}\right]^{1/2}. \tag{5.9}$$

The respective critical Ohnesorge numbers and their ratio are plotted as a function of λ in figures 1 and 2. The two individual critical values are increasing functions of λ, and the ratio $(Oh_c^{N2})/(Oh_c^{N1})$ again increases monotonically from a value of about 1.06 for a bubble $(\lambda=0)$, asymptoting to a value of about 1.51 in the limit $\lambda \to \infty$. In terms of the critical Reynolds numbers, this would imply that $Re_c^{N1} > Re_c^{N2}$ for all viscosity ratios. Thus, the theory predicts that an emulsion will have a negative N_1 and a positive

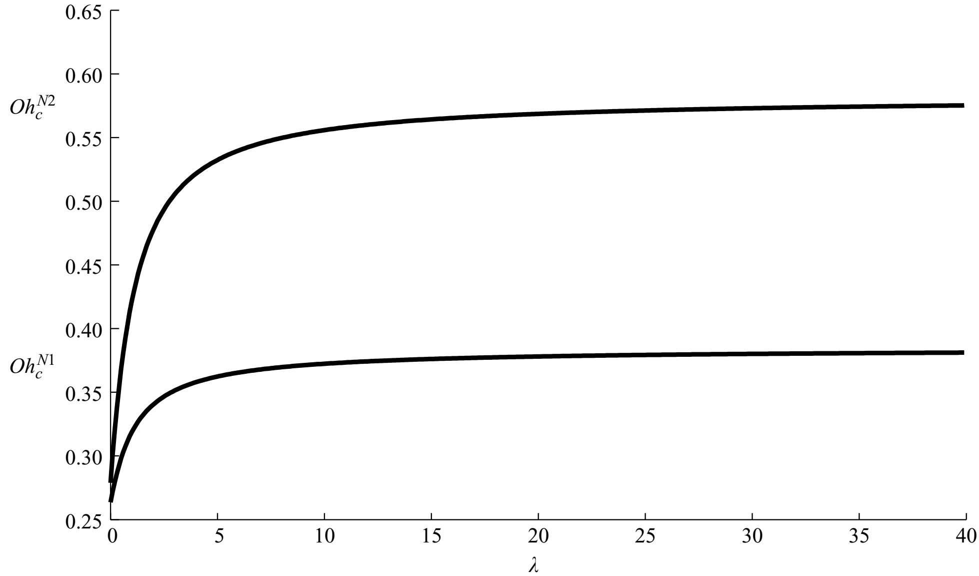

FIGURE 1. Oh_c^{N1} (lower curve) and Oh_c^{N2} (upper curve) as a function of the viscosity ratio λ, where Oh_c^{N1} and Oh_c^{N2} are the critical Reynolds numbers for a reversal in sign of N_1 and N_2, respectively.

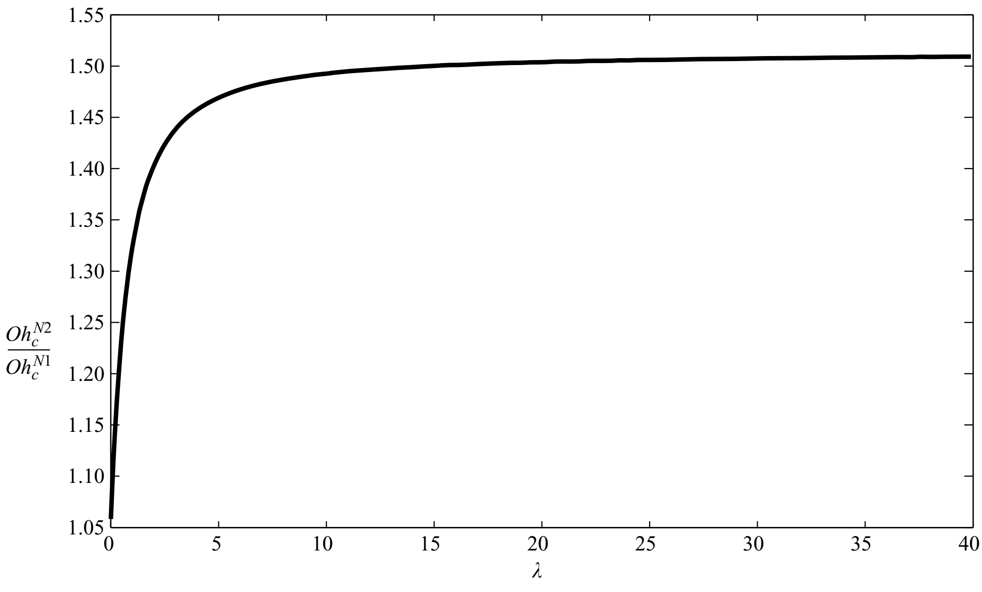

FIGURE 2. The ratio Oh_c^{N2}/Oh_c^{N1} as a function of λ.

N_2 for $Oh < Oh_c^{N1}(\lambda)$, a positive N_1 and a positive N_2 for $Oh_c^{N1}(\lambda) < Oh < Oh_c^{N2}(\lambda)$ and, finally, a positive N_1 and a negative N_2 for $Oh > Oh_c^{N2}(\lambda)$. The above predictions are, of course, valid only when the corresponding Reynolds and capillary numbers are still smaller than unity.

As in § 3, the limit $\lambda \to \infty$ serves as a check on the above predictions. In this limit, the contributions that do not explicitly involve the interfacial tension T (or Ca

in dimensionless terms) must reduce to those for a solid particle. Thus, the Taylor prediction for the $O(\phi)$ enhancement of the emulsion viscosity, $\{(5\lambda+2)/2(\lambda+1)\}\phi$, does reduce to the well-known Einstein correction in this limit; see (3.12) and (3.20). This must also apply to the $O(\phi Re)$ contributions to the normal stress differences in (5.3) and (5.4). In the limit of very viscous drops, one finds

$$\lim_{\lambda\to\infty} N_1 \approx -\frac{4}{3}\,\phi Re + O(\phi Ca), \tag{5.10}$$

$$\lim_{\lambda\to\infty} N_2 \approx \frac{2}{3}\,\phi Re + O(\phi Ca). \tag{5.11}$$

These are the expressions found originally by Lin *et al.* (1970) for a suspension of rigid spherical particles in simple shear flow in the limit of small but finite Re. In order to recover the result for a rigid particle suspension, one must therefore take the limit $\lambda, T \to \infty\,(Oh \to 0)$ corresponding to a very viscous drop with an infinitely stiff interface. The limit of a large drop viscosity alone is not necessarily equivalent to the rigid particle limit when one looks at steady-state rheological properties because the small outer viscosity, acting over a very long interval of time (which increases in proportion to the drop viscosity $\hat{\mu}$), acts to eventually induce a deformation that is independent of $\hat{\mu}$, provided the interfacial tension remains finite. In the limit $\lambda, T \to \infty$, however, only the $O(\phi Re)$ contributions survive, and as seen from (5.10) and (5.11), an inertial suspension in the dilute limit has a negative N_1 and a positive N_2. As mentioned earlier, unlike a dilute emulsion, a dilute suspension of rigid particles, in the absence of hydrodynamic interactions, exhibits a non-Newtonian rheology only for $Re \neq 0$.

An alternate interpretation of the aforementioned rigid particle limit allows us to partially extend our findings to the case of surfactant-covered drops. The relevant limit is $Ma \to \infty$, $Ca \to 0$ with $E = MaCa \approx O(1)$, where $Ma = [((\mathrm{d}T)/(\mathrm{d}c_i))\,c_{eq}]/a\mu\dot{\gamma}$ is the Marangoni number and E, the surfactant elasticity parameter, is a purely physicochemical quantity independent of the flow; here, c_{eq} is the equilibrium interfacial surfactant concentration, and $\mathrm{d}T/\mathrm{d}c_i$ is a measure of the sensitivity of interfacial tension to variations in the interfacial concentration c_i at $c_i = c_{eq}$. The above limit is typically realized for high molecular-weight surfactants for a sufficiently high surface coverage; the time scale for adsorption/de-sorption processes is usually much longer than the flow time scale for such surfactants, and the role of bulk processes in surfactant transport is therefore negligible. The interface in the limit $Ma \to \infty$ is infinitely sensitive to variations in the interfacial surfactant concentration and, therefore, supports a velocity field that is nearly solenoidal with a nearly uniform surfactant concentration. The problem of determining the rheology at $O(\phi Re)$ in the limit $Ca \to 0$, $Ma \to \infty$ reduces again to the solution of the governing equations of motion to $O(Re)$ in the exterior fluid, but with an imposed surface-solenoidal velocity boundary condition at the drop surface. Since, to $O(Ca)$, the drop may be treated as a sphere, the only interfacial velocity condition consistent with the symmetry of the ambient simple shear, and the additional constraint of being solenoidal, is a solid-body rotation field. Thus, the rheological problem in the limit reduces to that of a suspension of rigid spherical particles, and (5.10) and (5.11) may therefore be interpreted as the normal stress differences arising at $O(\phi Re)$ in a dilute emulsion of surfactant-laden drops in the limit $Ca \to 0$, $Ma \to \infty$. There are, again, contributions to both N_1 and N_2 at $O(\phi Ca)$, and these, of course, differ from those encountered at the beginning of this section. The normal stress differences given by (5.1) and (5.2) arise from the $O(Ca^2)$ deformation of a surfactant-free drop with a large but finite interfacial tension. On the other hand, the $O(\phi Ca)$ contributions referred to above are relevant to a surfactant-laden drop and arise because of the coupled effects of an $O(Ca^2)$ perturbation in the drop shape and an $O(Ma^{-1})$ variation in the (interfacial) surfactant concentration; the elastic interface in this case can support a finite shear stress. The rheology of a dilute emulsion in this latter regime has been determined by Vlahovska, Loewenberg & Blawzdziewicz (2005) for a viscosity ratio of unity via a perturbation analysis in the inertialess limit; they find the following expressions for N_1 and N_2 to $O(\phi Ca)$:

$$\lim_{Ma\to\infty,\lambda=1} N_1^{(0)} = \phi Ca\,\frac{5(4E+1)}{2E}, \tag{5.12}$$

$$\lim_{Ma\to\infty,\lambda=1} N_2^{(0)} = -\phi Ca\,\frac{5(13E+7)}{28E}. \tag{5.13}$$

As before, the shear viscosity of the emulsion remains unaltered at this order. Combining the expressions (5.10)–(5.13), one obtains, to $O(\phi Re)$, the following expressions for the first and second normal stress differences in a dilute emulsion of surfactant-laden drops with highly elastic interfaces in the limit of weak inertia and a viscosity ratio of unity:

$$\lim_{Ma\to\infty,\lambda=1} N_1 = \phi Ca\,\frac{5(4E+1)}{2E} - \frac{4}{3}\phi Re, \tag{5.14}$$

$$\lim_{Ma\to\infty,\lambda=1} N_2 = -\phi Ca\,\frac{5(13E+7)}{28E} + \frac{2}{3}\phi Re. \tag{5.15}$$

Since the analysis of Vlahovska *et al.* (2005) is only valid for $E\sim O(1)$, one again expects a change in the sign of both normal stress differences when $Re\sim O(Ca)$. The relevant critical Ohnesorge numbers are $Oh_c^{N1(Ma)} = [(8E)/\{15(4E+1)\}]^{1/2}$ and $Oh_c^{N2(Ma)} = [(56E)/\{15(13E+7)\}]^{1/2}$, and for $O(1)$ values of E, $Oh_c^{N2(Ma)}$ remains greater than $Oh_c^{N1(Ma)}$. Thus, the sequence of changes in rheological properties with increasing drop size remains the same as that outlined earlier in the introduction for the case of a dilute emulsion of surfactant-free drops. Although the case of a unit viscosity ratio is significantly easier to analyse in that the boundary integral representation of the disturbance velocity field in this case only involves a single layer potential (Pozrikidis 1992), one only expects a quantitative alteration in the rheology for other viscosity ratios. Furthermore, although the variation in the rheological characteristics of a dilute emulsion with *Ma* is non-monotonic and the analysis of Vlahovska *et al.* (2005) is only valid in the limit $Ma\to\infty$, one again expects the underlying physics and the general effect of inertia to remain qualitatively unaltered for $O(1)$ values of *Ma*. In summary, even for a dilute emulsion of surfactant-laden drops where the interfaces possess significant elasticity and, in addition, there are significant variations in the interfacial concentration, one expects a similar qualitative change in the rheology with inertial effects becoming increasingly important.

Moving on to the case of a steady extensional flow ($\boldsymbol{\Omega}=\mathbf{0}$), one finds that the $O(\phi Re)$ correction is identically zero for any λ. Thus, the bulk stress in a dilute emulsion subject to a steady extensional flow is given by

$$\Sigma_{ij} = -p_t\delta_{ij} + \mu\left\{2E_{ij} + 2\phi\frac{(5\lambda+2)}{2(\lambda+1)}E_{ij} + \phi Ca\left[\frac{3(19\lambda+16)(25\lambda^2+41\lambda+4)}{140(\lambda+1)^3}\right.\right.$$
$$\left.\left.\times\left\{E_{ik}E_{kj} - \frac{1}{3}\delta_{ij}(E_{kl}E_{kl})\right\}\right] + O(\phi^2, \phi Ca^2, \phi Re^{3/2})\right\}, \tag{5.16}$$

where the first effects of inertia now enter only at $O(\phi Re^{3/2})$. The Trouton ratio, defined as the ratio of the uniaxial extensional viscosity to the zero-shear viscosity, continues to be given by its inertialess approximation. The absence of any net effect, at $O(\phi Re)$, on the Trouton ratio is because the negative contribution due to the direct inertial stresses is cancelled by the positive $O(Re)$ correction to the stresslet integral. The sign of the latter contribution is consistent with the earlier results of Ramaswamy & Leal (1997), who found that weak inertial effects ($Re \leqslant O(1)$) contribute to an increase in the drop deformation in uniaxial extension at fixed Ca. This is because the deformation at the smaller values of Re is controlled by viscous stresses rather than the dynamic pressure contributions at the stagnation points; the latter retard drop elongation eventually leading to barrel-shaped drops at the highest Reynolds numbers and lowest viscosity ratios. Thus, rather counter-intuitively, although the drop deformation at $O(\phi Re)$ is expected to yield a more elongated shape, and thereby an increased tensile component of the surface tension forces along the extensional axis, the total inertial contribution to the Trouton ratio at this order is zero. Now, an inertial suspension must exhibit an extensional thickening rheology, this being an immediate consequence of Helmholtz's minimum dissipation theorem (see Batchelor 1967; Kim & Karrila 1991). Owing to the absence of vorticity, a rigid particle in an ambient extensional flow will not rotate at any Re, and the disturbance velocity field must therefore satisfy the same boundary condition independent of Re, a requirement for the applicability of the aforementioned theorem. This has already been pointed out by Ryskin (1980), who determined the Trouton viscosity of a rigid particle suspension for a range of Re. Since the first effects of inertia in extensional flow enter at $O(\phi Re^{3/2})$, the above conclusion must, in fact, also hold for a dilute emulsion; in particular, the $O(\phi Re^{3/2})$ contribution to the Trouton ratio is likely to have an extensional-thickening character for any viscosity ratio. The generalization becomes possible on account of the similar behaviour of drops and particles at $O(Re^{3/2})$—both act as point-force dipoles on length scales that contribute dominantly to the bulk stress (Subramanian & Koch 2010). Ryskin (1980), in fact, also calculated the extensional viscosities for drops (both inviscid and those having the same viscosity as the continuous phase) and found the behaviour to be qualitatively the same as that for rigid particles, viz. a monotonic increase with Re. The range of small but finite Re was, however, not resolved well enough in the calculations for one to extract a scaling behaviour. The shear thickening of an inertial suspension predicted in a vortical flow such as simple shear (Lin *et al.* 1970) is not an obvious consequence. Unlike an extensional flow, the particle angular velocity and therefore the velocity boundary conditions are a function of Re in this case.

Finally, we compare our theoretical predictions for simple shear flow with the recent simulation results of Li & Sarkar (2005), who first emphasized the important role of inertia in emulsion rheology and were able to relate the effects of inertia on drop deformation to corresponding changes in the bulk stress. The authors studied the effect of inertia on the deformation of a single drop and the resulting steady shear rheology of a dilute emulsion of such drops. The dilute emulsion was obtained by simulating a single drop in simple shear flow generated by two infinite parallel plates separated vertically and moving in opposite directions, with periodic boundary conditions in the two horizontal (flow and vorticity) directions. The Navier–Stokes equations were solved using a front-tracking finite-difference method with the singular interfacial tension forces being distributed over a thin layer surrounding the drop. The viscosity ratio was taken as unity in the plots for the rheological properties. As seen from the expression (3.5) for the bulk stress, one only has contributions from the

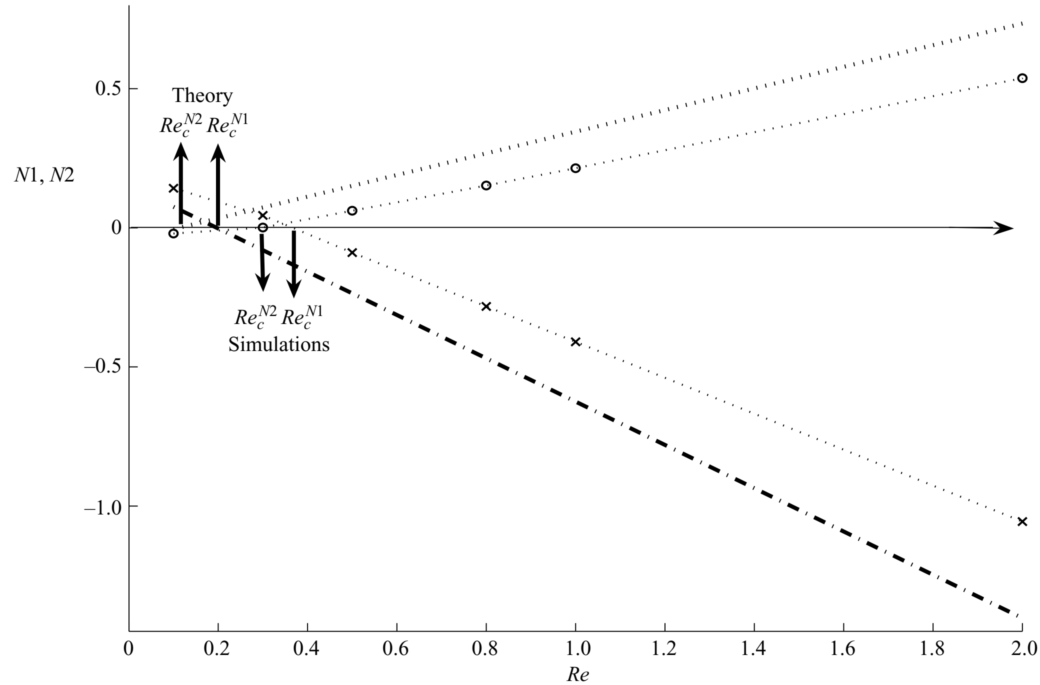

FIGURE 3. The variation of the first and the second normal stress differences, from both theory (equations (5.3) and (5.4)) and simulation, as a function of Re for $Ca = 0.02$, $\lambda = 1$. The bold dotted and dash-dotted lines denote the theoretical predictions, while the symbols denote the simulation results.

interfacial and inertial stresses for this viscosity ratio. The simulations were carried out for a range of Reynolds numbers from 0.1 to 3 and for capillary number ranging from 0.02 to about 0.1. As already seen in earlier sections, the theory predicts the shear viscosity to remain unchanged to $O(\phi Re)$; thus, a dilute emulsion, to $O(\phi Re)$, will exhibit a shear-thinning behaviour for the same reason as that for an inertialess emulsion, this being related to the increasing alignment of the deformed drop with the flow direction in the absence of inertia; in the limit of small Ca, the first effects of shear thinning appear at $O(\phi Ca^2)$ (see Barthes Biesel & Acrivos 1973) and have not been included in the analysis here. In simulations, we not only observe shear thinning at smaller values of Re, but also observe shear thickening at larger values of Re of order unity. Since a dilute emulsion always shear thins in the absence of inertia, the observed shear thickening is likely a result of inertia. As indicated above, for small Re, an inertial contribution of this nature arises at $O(\phi Re^{3/2})$ and is analysed in detail in a forthcoming publication (Subramanian & Koch 2010). Herein, we restrict the comparison with the $O(\phi Re)$-accurate theory to normal stress differences.

In figures 3 and 4, we plot N_1 and N_2 as a function of Re for $Ca = 0.02$ and $Ca = 0.05$, respectively. The qualitative agreement between the theoretical predictions and the simulation results is readily seen. In both cases, the normal stress differences change sign with increasing Re, and simulations predict Re_c^{N1} to be greater than Re_c^{N2}, in agreement with theory. Furthermore, the observed drop deformation on account of inertial stresses is again consistent with theoretical predictions, viz. the tilting towards the velocity gradient axis. However, the comparison also reveals a quantitative

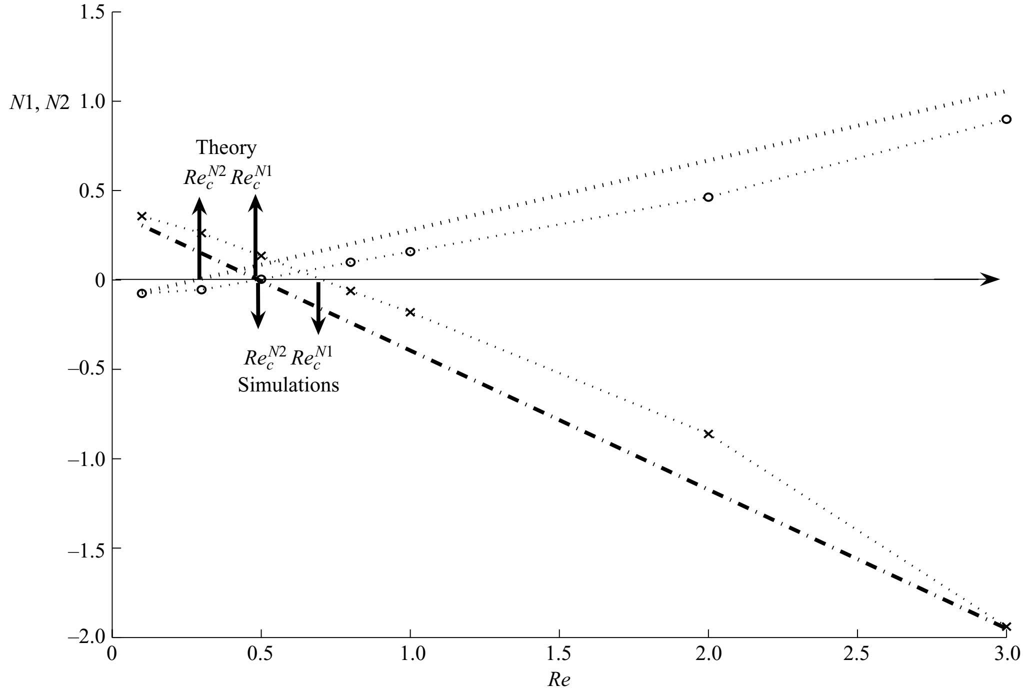

FIGURE 4. The variation of the first and the second normal stress differences, from both theory (equations (5.3) and (5.4)) and simulation, as a function of Re for $Ca = 0.05$, $\lambda = 1$. The bold dotted and dash-dotted lines denote the theoretical predictions, while the symbols denote the simulation results.

discrepancy that persists down until the smallest Reynolds numbers ($Re \approx 0.1$). Specifically, the critical Reynolds numbers predicted by theory for both N_1 and N_2 are always smaller than those found in the simulations. Since the theoretical and simulation curves are nearly parallel to one another and the difference is in the intercepts on the Re axis, the initial suggestion is that of a discrepancy independent of Re. The likely candidate, for small Ca, is thus the $O(\phi Ca^2)$ correction referred to above. Based on the qualitative discussion therein, one expects the $O(\phi Ca^2)$ correction to further increase N_1 and decrease N_2; these changes are indeed in the right direction for the theory to approach the numerics. The analysis for a neutrally buoyant drop to this order was originally carried out by Barthes Biesel & Acrivos (1973) and, more recently, by Greco (2002); neither analysis, however, proceeds to calculate the rheological properties from the expressions for the $O(Ca^2)$ velocity and pressure fields. An $O(Ca^2)$ rheological calculation is clearly beyond the scope of the present analysis. Nevertheless, it is worth noting that the capillary numbers involved in figures 3 and 4 are very small, and the required shift in the theory curves, for agreement with simulation, corresponds to a numerical pre-factor, at $O(Ca^2)$, of about a 100 for a viscosity ratio of unity—a rather unlikely scenario. A closer examination, however, reveals a slight divergence between the numerical and theoretical curves with increasing Re. Now, it may be shown that the contributions to N_1 and N_2 at $O(\phi Re^{3/2})$ have the same sign as those at $O(\phi Ca^2)$ (Subramanian & Koch 2010). Furthermore, for the range of Re and Ca examined in the simulations, the $O(Re^{3/2})$ correction is expected to be significantly larger than that at $O(Ca^2)$. Thus, it is likely that the above discrepancy may, at least in part, be because of the neglect of higher-order inertial corrections in the current analysis.

6. Conclusions

In this paper, we have derived the stress in a dilute emulsion of neutrally buoyant drops with small but non-zero Reynolds number $Re = \dot{\gamma} a^2 \rho / \mu$ and capillary number $Ca = \mu a \dot{\gamma} / T$, when the drop radius a is small compared with the length scale L of the imposed flow field. In § 2 and 3, we derived expressions for the fluid velocity and pressure fields, and the disperse phase stress, to $O(Re)$, when the drops are suspended in an ambient linear flow without vortex stretching; the absence of vortex stretching may either be on account of the vorticity vector being orthogonal to the rate of strain tensor as in a planar linear flow or because the vorticity is identically zero as is the case in an extensional flow. The stress for a general three-dimensional linear flow was derived in § 4 using the reciprocal theorem. Unlike a rigid particle, this analysis required two applications of the reciprocal theorem—one to determine the required moment of the surface force density and the other to determine the moment of the tangential velocity on the drop surface that also contributes to the drop stresslet. The inclusion of contributions corresponding to a weak nonlinearity in the ambient flow field and temporal variations of the ambient strain rate, both of which contribute to the stress at $O(Re)$, then led to the expression (4.36) for the disperse phase stress tensor which constitutes the primary result of the paper.

The application of this rheological result would require knowledge of the drop volume fraction and its possible dependence on position. It is well known that deformable and/or finite Reynolds number drops can migrate because of nonlinearities in the imposed flow field or the presence of walls (Taylor 1932; Ho & Leal 1974; Vasseur & Cox 1976; Schonberg & Hinch 1989). When Ca, Re and a/L are all small, as assumed in the present study, the migration velocity is small compared with the mean velocity of the suspension. This suggests that spatial variations in the volume fraction may be small in Lagrangian unsteady flows as long as a mechanism is available to produce an initial uniform drop distribution at the inlet to the flow cell. The most pronounced variations in drop volume fraction may be expected in unidirectional flows or nearly unidirectional flows where drops experience the same flow conditions over a sufficient time to migrate an appreciable distance.

One useful approach to extend our knowledge of the rheology of finite Reynolds number drops would be to simulate the planar Couette flow of an emulsion. For example, the front-tracking finite-difference method of Esmaeeli & Tryggvason (1998) could be used to simulate a suspension of slightly deformable, neutrally buoyant drops bounded by two planar walls over a range of Reynolds numbers and volume fractions. The shear stress and first normal stress difference could be determined directly from the force on the walls, while more detailed insights into the various contributions to the stress could be obtained using (3.2). Such a study could determine the range of conditions over which the present predictions are accurate as well as revealing the changes in rheology that arise at finite Reynolds number and volume fraction. The planar Couette geometry would lead to a Lagrangian steady mean suspension velocity, allowing an extended period for (transverse) drop migration to occur. Nonetheless, one could expect the wall-induced migration of drops to be balanced by hydrodynamic diffusion. At steady state where no net flux occurs, such a balance may be written as

$$V(y)\phi - D(\phi, y)\frac{d\phi}{dy} = 0, \tag{6.1}$$

where $V(y)$ is the migration velocity, $D(\phi, y)$ the shear-induced gradient diffusivity and y is the coordinate normal to the wall. In the case of a dilute suspension of rigid particles, Vasseur & Cox (1976) have determined V as a function of y. Recent numerical simulations indicate a reasonably small variation of volume fraction across the Couette cell (Wang *et al.* 2009), thereby allowing for an interpretation in terms of a local rheological response based on a homogeneously distributed disperse phase.

In summary, our rheological equation for the disperse phase stress in a dilute emulsion (4.36) is applicable in the limits $a/L \ll 1$, $Re \ll 1$, $Ca \ll 1$ and $\phi \ll 1$. In simple shear flow it yields first and second normal stress differences which change sign depending on the value of the Ohnesorge number $Oh = (Ca/Re)^{1/2}$. This constitutive equation should be applied in conjunction with a mass balance determining the spatial dependence of the drop volume fraction. However, there may be many situations in which the variations in the volume fraction are modest.

Appendix A. Velocity field constants

The constants c_i appearing in the expressions (2.19) and (2.20) for the exterior velocity and pressure fields are given below:

$$c_1 = \frac{5005\lambda^3 + 7722\lambda^2 + 2288\lambda + 112}{1144(\lambda+1)^2},$$

$$c_2 = \frac{5\lambda^2 + 2\lambda}{\lambda + 1},$$

$$c_3 = \frac{19305\lambda^3 + 29172\lambda^2 + 11440\lambda + 1032}{10296(\lambda+1)^2},$$

$$c_4 = \frac{(5\lambda+2)^2}{4(\lambda+1)},$$

$$c_5 = \frac{5\lambda+2}{2},$$

$$c_6 = \frac{42042\lambda^4 + 177177\lambda^3 + 204204\lambda^2 + 76996\lambda + 6304}{41184(2\lambda+5)(\lambda+1)^2},$$

$$c_7 = \frac{\lambda}{2},$$

$$c_8 = \frac{35178\lambda^4 + 132561\lambda^3 + 133276\lambda^2 + 41532\lambda + 4016}{41184(2\lambda+5)(\lambda+1)^2},$$

$$c_9 = \frac{426426\lambda^4 + 1728441\lambda^3 + 1942512\lambda^2 + 735778\lambda + 90412}{144144(2\lambda+5)(\lambda+1)^2},$$

$$c_{10} = \frac{3\lambda^2 + 3\lambda + 1}{9(\lambda+1)},$$

$$c_{11} = \frac{234234\lambda^4 + 959673\lambda^3 + 1077648\lambda^2 + 383426\lambda + 26348}{144144(2\lambda+5)(\lambda+1)^2},$$

$$c_{12} = \frac{282282\lambda^4 + 1134705\lambda^3 + 1268124\lambda^2 + 480828\lambda + 63504}{288288(2\lambda+5)(\lambda+1)^2},$$

$$c_{13} = \frac{138138\lambda^4 + 582153\lambda^3 + 675532\lambda^2 + 248596\lambda + 15456}{288288(2\lambda+5)(\lambda+1)^2},$$

$$c_{14} = \frac{\lambda^2}{4(\lambda+1)},$$

$$c_{15} = \frac{3\lambda^2 + 3\lambda + 4}{18(\lambda+1)},$$

$$c_{17} = \frac{1075074\lambda^4 + 4448301\lambda^3 + 5107388\lambda^2 + 1947684\lambda + 205408}{1441440(2\lambda+5)(\lambda+1)^2},$$

$$c_{18} = \frac{1027026\lambda^4 + 4135989\lambda^3 + 4610892\lambda^2 + 1699436\lambda + 189392}{1441440(2\lambda+5)(\lambda+1)^2}.$$

The constants c_i' appearing in the expressions (2.21) and (2.22) for the interior velocity and pressure fields are given below:

$$\left.\begin{aligned}
c_1' &= \frac{7}{286},\\
c_2' &= \frac{2145\lambda^3 + 3718\lambda^2 + 528\lambda - 504}{20592(\lambda+1)},\\
c_3' &= \frac{15015\lambda^3 + 30602\lambda^2 + 12848\lambda + 1048}{82368(\lambda+1)},\\
c_4' &= \frac{48906\lambda^4 + 191477\lambda^3 + 242606\lambda^2 + 108500\lambda + 16580}{82368(2\lambda+5)(\lambda+1)},\\
c_5' &= \frac{15015\lambda^3 + 21450\lambda^2 - 5456\lambda - 8104}{82368(\lambda+1)},\\
c_6' &= \frac{6006\lambda^4 + 9867\lambda^3 + 41106\lambda^2 + 74540\lambda + 29180}{82368(2\lambda+5)(\lambda+1)},\\
c_7' &= \frac{2145\lambda^3 + 3718\lambda^2 + 8536\lambda + 7504}{192192(\lambda+1)},\\
c_8' &= \frac{282282\lambda^4 + 1063205\lambda^3 + 1097174\lambda^2 + 361896\lambda + 53760}{576576(2\lambda+5)(\lambda+1)},\\
c_9' &= -\frac{34034\lambda^4 + 115401\lambda^3 + 104078\lambda^2 + 7496\lambda - 17920}{192192(2\lambda+5)(\lambda+1)},\\
c_{10}' &= \frac{2145\lambda^3 + 4862\lambda^2 - 3190\lambda - 5366}{82368(\lambda+1)},\\
c_{11}' &= \frac{534534\lambda^4 + 1987843\lambda^3 + 1893814\lambda^2 + 524990\lambda + 76370}{576576(2\lambda+5)(\lambda+1)},\\
c_{12}' &= \frac{516\lambda^2 + 156\lambda - 7}{2016},\\
c_{13}' &= \frac{2145\lambda^3 + 2574\lambda^2 - 7766\lambda - 7654}{82368(\lambda+1)},
\end{aligned}\right\} \tag{A 1}$$

$$\left.\begin{aligned}
c'_{14} &= -\frac{234234\lambda^4 + 830973\lambda^3 + 636714\lambda^2 - 204670\lambda - 236530}{576576(2\lambda+5)(\lambda+1)}, \\
c'_{15} &= -\frac{156\lambda^2 + 12\lambda - 49}{2016}, \\
c'_{16} &= \frac{1}{11}, \\
c'_{17} &= \frac{10725\lambda^3 + 18590\lambda^2 - 5784\lambda - 10944}{7488(\lambda+1)}, \\
c'_{18} &= \frac{10725\lambda^3 + 31694\lambda^2 + 28848\lambda + 10584}{52416(\lambda+1)}, \\
c'_{19} &= \frac{276\lambda^2 + 100\lambda - 19}{144}, \\
c'_{20} &= \frac{10725\lambda^3 + 5486\lambda^2 - 23568\lambda - 15624}{52416(\lambda+1)}, \\
c'_{21} &= \frac{8\lambda^2 - 3}{8}, \\
c'_{22} &= -\frac{60\lambda^2 - 44\lambda - 37}{144}, \\
c'_{23} &= \frac{2145\lambda^3 + 3718\lambda^2 - 9300\lambda - 10332}{52416(\lambda+1)}, \\
c'_{24} &= \frac{20\lambda^2 + 8\lambda + 7}{48}, \\
c'_{25} &= \frac{2145\lambda^3 + 3718\lambda^2 - 1474\lambda - 2506}{82368(\lambda+1)}, \\
c'_{26} &= \frac{2145\lambda^3 + 3718\lambda^2 - 9482\lambda - 10514}{82368(\lambda+1)}, \\
c'_{27} &= \frac{558558\lambda^4 + 1755039\lambda^3 + 203814\lambda^2 - 1411082\lambda - 458990}{2882880(2\lambda+5)(\lambda+1)}, \\
c'_{28} &= \frac{942942\lambda^4 + 4029311\lambda^3 + 6081686\lambda^2 + 5059382\lambda + 2023490}{2882880(2\lambda+5)(\lambda+1)}, \\
c'_{29} &= \frac{1680\lambda^3 + 1452\lambda^2 + 1180\lambda + 273}{10080}, \\
c'_{30} &= -\frac{1680\lambda^3 + 3252\lambda^2 + 1900\lambda + 483}{10080}.
\end{aligned}\right\} \quad \text{(A 1, cont.)}$$

Appendix B. Expression for the surface force density: $\boldsymbol{\sigma} \cdot \boldsymbol{n}\,|_{r=1}$

The expression for the surface force density evaluated using the exterior velocity and pressure fields is given by

$$\left.\begin{aligned}
\boldsymbol{\sigma} \cdot \boldsymbol{n}\,|_{r=1} &= \left(-\frac{13a_1}{2} - \frac{15a_2^2}{16} + \frac{5a_3}{2} + \frac{5a_3^2}{8} - 12A - 7C - 7E - 7A'\right)(\boldsymbol{\Gamma} : \boldsymbol{nn})^2\boldsymbol{n} \\
&+ \left(\frac{19a_1}{16} + \frac{a_2^2}{6} + \frac{3a_3}{4} - \frac{a_2}{6} - 3C + \frac{8A}{3} + 2E + A'\right)(\boldsymbol{\Gamma} : \boldsymbol{nn})(\boldsymbol{\Gamma} \cdot \boldsymbol{n})
\end{aligned}\right\} \quad \text{(B 1)}$$

$$
\left.
\begin{aligned}
&+\left(\frac{19a_1}{16}+\frac{a_2^2}{6}-\frac{3a_3}{4}-\frac{a_2}{12}+2C+\frac{8A}{3}-3E+A'\right)(\boldsymbol{\Gamma}:\boldsymbol{nn})(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{n})\\
&+\left(\frac{15a_1}{16}+\frac{5a_2^2}{16}+\frac{3a_3}{2}+\frac{a_3^2}{8}-8G+\frac{4A}{3}+E-4C'+\frac{6A'}{7}-3O\right)(\boldsymbol{\Gamma}\cdot\boldsymbol{n})\cdot(\boldsymbol{\Gamma}\cdot\boldsymbol{n})\boldsymbol{n}\\
&+\left(\frac{15a_1}{16}+\frac{5a_2^2}{16}-\frac{a_2}{12}+\frac{a_3^2}{8}-8K+\frac{4A}{3}+C-4E'+\frac{6A'}{7}-3M\right)(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{n})\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{n})\boldsymbol{n}\\
&+\left(\frac{15a_1}{8}+\frac{5a_2^2}{8}+\frac{3a_3}{2}-\frac{a_2}{12}+\frac{a_3^2}{4}-8I+\frac{8A}{3}+C+E-4G'+\frac{12A'}{7}-3Q-3S\right)\\
&\quad\times(\boldsymbol{\Gamma}\cdot\boldsymbol{n})\cdot(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{n})\boldsymbol{n}\\
&+\left(-\frac{5a_1}{16}-\frac{a_2^2}{12}-\frac{a_3}{2}+\frac{a_2}{12}+\frac{16G}{5}+C'-\frac{8A}{21}-O+\frac{3E}{5}-\frac{A'}{7}\right)(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{\Gamma}\cdot\boldsymbol{n})\\
&+\left(-\frac{5a_1}{16}-\frac{a_2^2}{12}+\frac{16K}{5}+E'-\frac{8A}{21}-M+\frac{3C}{5}-\frac{A'}{7}\right)(\boldsymbol{\Gamma}\cdot\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{n}),\\
&+\left(-\frac{5a_1}{16}-\frac{a_2^2}{12}-\frac{a_3}{4}+\frac{8I}{5}+S-2Q-\frac{8A}{21}+\frac{4C}{5}-\frac{E}{5}+\frac{G'}{2}-\frac{A'}{7}\right)(\boldsymbol{\Gamma}\cdot\boldsymbol{\Gamma}\cdot\boldsymbol{n}),\\
&+\left(-\frac{5a_1}{16}-\frac{a_2^2}{12}-\frac{a_3}{4}+\frac{a_2}{12}+Q-2S+\frac{8I}{5}-\frac{8A}{21}+\frac{4E}{5}-\frac{C}{5}+\frac{G'}{2}-\frac{A'}{7}\right)(\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{\Gamma}^\dagger\cdot\boldsymbol{n})\\
&+\left(-\frac{5a_1}{24}-\frac{a_2^2}{12}+\frac{8I}{5}-\frac{4A}{21}-4U-\frac{4A'}{35}+\frac{2G'}{3}\right)(\boldsymbol{\Gamma}:\boldsymbol{\Gamma})\boldsymbol{n}\\
&+\left(-\frac{5a_1}{24}-\frac{a_2^2}{12}+\frac{8(G+K)}{5}-\frac{4A}{21}-4W-\frac{4A'}{35}+\frac{2(C'+E')}{3}\right)(\boldsymbol{\Gamma}:\boldsymbol{\Gamma}^\dagger)\boldsymbol{n}.
\end{aligned}
\right\}
\tag{B 1, cont.}
$$

The constants in the above stress equation are tabulated below:

$$
\left.
\begin{aligned}
A&=-\frac{5005\lambda^3+7722\lambda^2+2288\lambda+112}{4576(\lambda+1)^3},\\
A'&=-\frac{7(19305\lambda^3+29172\lambda^2+11440\lambda+1032)}{20592(\lambda+1)^3},\\
C&=\frac{42042\lambda^4+177177\lambda^3+204204\lambda^2+76996\lambda+6304}{41184(\lambda+1)^3(2\lambda+5)},\\
C'&=-\frac{3\lambda^2+3\lambda+4}{18(\lambda+1)^2},\\
E&=\frac{35178\lambda^4+132561\lambda^3+133276\lambda^2+41532\lambda+4016}{41184(\lambda+1)^3(2\lambda+5)},\\
E'&=\frac{2(3\lambda^2+3\lambda+1)}{(\lambda+1)^2},\\
G&=\frac{582582\lambda^4+2372799\lambda^3+2703844\lambda^2+1056708\lambda+144704}{(\lambda+1)^3(2\lambda+5)},\\
G'&=\frac{1}{2\lambda(\lambda+1)},\\
I&=\frac{195195\lambda^3+314028\lambda^2+134420\lambda+16128}{144144(\lambda+1)^3},
\end{aligned}
\right\}
\tag{B 2}
$$

$$\left.\begin{aligned}
I' &= -\frac{5\lambda + 2}{12(\lambda + 1)},\\
K &= \frac{198198\lambda^4 + 835263\lambda^3 + 974116\lambda^2 + 352004\lambda + 16576}{288288(\lambda + 1)^3(2\lambda + 5)},\\
K' &= -\frac{5\lambda + 2}{12(\lambda + 1)},\\
M &= \frac{\lambda}{12(\lambda + 1)},\\
O &= -\frac{\lambda}{12(\lambda + 1)},\\
Q &= \frac{1}{30},\\
S &= -\frac{1}{30},\\
U &= \frac{\lambda}{12(\lambda + 1)},\\
W &= \frac{\lambda}{12(\lambda + 1)},\\
a_1 &= \frac{5\lambda^2 + 2\lambda}{(\lambda + 1)^2},\\
a_2 &= \frac{5\lambda + 2}{(\lambda + 1)},\\
a_3 &= \frac{\lambda}{\lambda + 1}.
\end{aligned}\right\} \quad \text{(B 2, cont.)}$$

Appendix C. Solution for the flow driven by a stress jump across a spherical interface

Here, we solve for the flow driven in both the exterior and interior of a spherical drop across whose interface there is a specified jump in the tangential stress. Physically, such a jump in shear stress may be attributed to a particular (instantaneous) distribution of surfactant at the interface. Assuming all relevant length scales to be small enough for inertial forces to be negligible, we have the following governing equations:

$$\nabla^2 \tilde{\boldsymbol{u}}^+ - \nabla \tilde{p}^+ = \boldsymbol{0}, \tag{C 1}$$

$$\nabla \cdot \tilde{\boldsymbol{u}}^+ = 0, \tag{C 2}$$

in the exterior fluid, and

$$\nabla^2 \tilde{\boldsymbol{u}}^- - \nabla \tilde{p}^- = \boldsymbol{0}, \tag{C 3}$$

$$\nabla \cdot \tilde{\boldsymbol{u}}^- = 0, \tag{C 4}$$

in the interior fluid, with the following boundary conditions:

$$\tilde{\boldsymbol{u}}^+ = \tilde{\boldsymbol{u}}^-, \tag{C 5}$$

$$\tilde{\boldsymbol{u}}^+ \cdot \boldsymbol{n} = 0, \tag{C 6}$$

$$(\boldsymbol{\sigma}^+ \cdot \boldsymbol{n}) \cdot (\boldsymbol{I} - \boldsymbol{nn}) - \lambda(\boldsymbol{\sigma}^- \cdot \boldsymbol{n}) \cdot (\boldsymbol{I} - \boldsymbol{nn}) = (\hat{\boldsymbol{B}} \cdot \boldsymbol{n}) \cdot (\boldsymbol{I} - \boldsymbol{nn}), \tag{C 7}$$

at the interface $r = 1$, with the usual far-field decay conditions for $(\tilde{\boldsymbol{u}}^+, \tilde{\boldsymbol{\sigma}}^+)$ and the condition of regularity at the origin for $(\tilde{\boldsymbol{u}}^-, \tilde{\boldsymbol{\sigma}}^-)$. Using standard procedure involving an appropriate choice of spherical harmonic solutions (e.g. see Leal 1992), one finds

$$\tilde{\boldsymbol{u}}^+ = \frac{1}{(\lambda+1)}\left[\left(-\frac{3}{10r^5}+\frac{1}{2r^7}\right)\boldsymbol{r}(\hat{\boldsymbol{B}}:\boldsymbol{rr}) - \frac{1}{5r^5}\hat{\boldsymbol{B}}\cdot\boldsymbol{r}\right], \tag{C 8}$$

$$\tilde{\boldsymbol{u}}^- = \frac{1}{(\lambda+1)}\left[\left(\frac{3}{10}-\frac{r^2}{2}\right)\hat{\boldsymbol{B}}\cdot\boldsymbol{r} + \frac{1}{5}\boldsymbol{r}(\hat{\boldsymbol{B}}:\boldsymbol{rr})\right], \tag{C 9}$$

$$\tilde{p}^+ = -\frac{3}{5(\lambda+1)}\frac{\hat{\boldsymbol{B}}:\boldsymbol{rr}}{r^5}, \tag{C 10}$$

$$\tilde{p}^- = -\frac{21\lambda}{10(\lambda+1)}\hat{\boldsymbol{B}}:\boldsymbol{rr}, \tag{C 11}$$

for the velocity and pressure fields, and the corresponding jump in normal stress at $r=1$ is therefore found to be

$$(\tilde{\boldsymbol{\sigma}}^+ - \lambda\tilde{\boldsymbol{\sigma}}^-):\boldsymbol{nn} = -\frac{(9\lambda+6)}{10(\lambda+1)}\hat{\boldsymbol{B}}:\boldsymbol{nn}. \tag{C 12}$$

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