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Second-order theory for the deformation of a Newtonian drop in a stationary flow field

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The classical perturbative theory for a single drop immersed in a flowing immiscible fluid is revisited, with the well-known capillary number Ca , ratio of viscous to interfacial stresses, as the expansion parameter. Although the analysis is here limited to the Newtonian case under steady-state conditions, the perturbation method is innovative, as it makes use of rotational invariance to obtain workable tensorial representations of the pressure and velocity fields, and of the drop shape, at any order in Ca . The method is much less cumbersome than the classical one, based on an expansion in spherical harmonics. The analytical second-order solution thus obtained differs somewhat from previously reported results. © 2002 American Institute of Physics. [DOI: 10.1063/1.1445182]

I. INTRODUCTION

In this paper we reconsider the well-known problem of a fluid drop freely suspended in a second unbounded fluid that flows in a prescribed way at infinity. Both fluids are inertialess, Newtonian fluids. The study of such an idealized problem is directly relevant in dilute systems, where the dispersed phase is made up of drops far apart from each other. It should be emphasized that, even if the fluids are Newtonian, the resulting dispersed blend is non-Newtonian, because of elastic effects due to the presence of an interface.¹

The mathematical complexities of the drop problem are well described in several reviews.^{2,3} In inertialess situations, the major source of difficulty is in the fact that some of the boundary conditions to be satisfied, i.e., the matching of inner and outer velocities as well as a force balance involving interfacial tension, are assigned on the surface of the deforming drop, which is itself unknown. Pressure and velocity fields, inside and outside the drop, and drop shape must all be determined simultaneously, which is a very complex three-dimensional (3D) moving boundary problem. Lacking an analytic solution to the general problem, simulations have recently been developed,^{4,5} which however require considerable computational efforts.

Analytic results (see the reviews^{2,3}) have only been obtained under limiting conditions (of flow intensity and/or of constitutive properties) such that the drop deformation is small at all times. In fact, small drop deformations occur in two cases, either for small capillary number Ca (Ca is the ratio of viscous to interfacial stresses), or for very large values of the ratio λ of inner to outer viscosity. The case examined in this paper is the former one, for which Ca is the obvious expansion parameter (see Sec. II). The zeroth-order problem corresponds to statics, i.e., to the equilibrium spherical shape of the drop. To proceed at higher orders in the expansion, a standard “domain perturbation” technique

is used,⁶ whereby interfacial boundary conditions can be “transferred” to a fictitious spherical interface, in view of the smallness of the drop deformation (see Sec. III).

The first-order problem was solved long ago by Taylor,⁷ who used a general solution of the Navier–Stokes equations given by Lamb⁸ in terms of series of spherical harmonics. By adopting the same mathematical approach, several authors later attacked the second-order problem.^{9–14} It has to be mentioned, however, that the *complete* second-order solution inclusive of pressure and velocity fields has never been published, the only available results in the open literature being those for the drop shape.

In this paper we deal with the second-order problem for steady state situations only, but we will do so without using Lamb’s series of spherical harmonics. Instead, we present an analysis based on standard arguments of continuum mechanics, whereby rotational invariance¹⁵ is used to readily obtain the tensorial forms of all the unknown fields, and of the drop shape. Solving the balance equations and imposing the interfacial boundary conditions is then straightforward, and the complete solution is obtained after some lengthy but plain algebra.

The solution method chosen in this paper, though limited to steady-state situations, has the advantage of compactness and simplicity, and allows one to readily check all the intermediate calculations. Also (see Sec. V), a much more transparent condition for drop volume conservation is obtained. (Drop volume conservation admittedly generated problems in some of the previous calculations; see Refs. 9 and 10.) Concerning the comparison of our second-order results with existing predictions for the drop shape,^{9–13} we have found some discrepancies. These are briefly discussed in the final section of the paper, also showing how intricate is the history of the drop- Ca^2 -problem.

The paper is organized as follows. First, we state the general mathematical problem, and its nondimensionalized version, and illustrate the perturbation procedure. In Secs. IV and V, we obtain the above-mentioned tensorial form of the pressure and velocity fields, and of the drop shape, and dis-

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cuss drop volume conservation. The complete second-order (steady-state) solution is given in Sec. VI, and discussed in the last section. For convenience, Taylor first-order solution and various mathematical details are reported in the Appendices.

II. THE MATHEMATICAL PROBLEM

We consider the motion of a neutrally buoyant drop immersed in a fluid of infinite extent, subject to a given velocity gradient $\mathbf{L}^{(\infty)} \equiv \nabla \mathbf{V}^{(\infty)}$ “at infinity” (see Refs. 2 and 3). The two fluids are taken to be mutually immiscible. In isothermal conditions, the motion of both fluids is governed by the equations of mass and momentum balance. For the constant-density, inertialess case considered here, and for Newtonian fluids, we then have the continuity and Navier–Stokes equations (we report well-known equations in detail with the purpose of defining the symbols used)

$$\begin{aligned} \nabla \cdot \mathbf{V} &= 0, \\ -\nabla P + \eta \nabla^2 \mathbf{V} &= \mathbf{0}, \\ \nabla \cdot \hat{\mathbf{V}} &= 0, \\ -\nabla \hat{P} + \hat{\eta} \nabla^2 \hat{\mathbf{V}} &= \mathbf{0}, \end{aligned} \quad (1) \quad (\hat{1})$$

for the external and internal fields, respectively. (From now on, all quantities referring to the interior fluid will be denoted with carets.)

The boundary conditions are assigned at the interface, and at infinity. By describing the drop surface at time t as $F(\mathbf{r}, t) = 0$ (\mathbf{r} is the position vector of a point at the interface, while the generic position vector is denoted by \mathbf{R}), the external normal to the drop surface is $\mathbf{N} = \nabla F / |\nabla F|$. The interfacial boundary conditions then read¹⁶

$$\mathbf{V} = \hat{\mathbf{V}}, \quad (2)$$

$$\mathbf{V} \cdot \mathbf{N} = \frac{1}{|\nabla F|} \frac{\partial F}{\partial t}, \quad (3)$$

$$\sigma \mathbf{N} \nabla \cdot \mathbf{N} = (\hat{P} - P) \mathbf{N} + 2(\eta \mathbf{D} - \hat{\eta} \hat{\mathbf{D}}) \cdot \mathbf{N} \quad (4)$$

with $\mathbf{D} \equiv (\mathbf{L} + \mathbf{L}^T)/2 = (\nabla \mathbf{V} + \nabla \mathbf{V}^T)/2$ the local strain rate at the interface, and σ the interfacial tension. Boundary conditions at infinity are simply

$$\mathbf{V} \rightarrow \mathbf{L}^{(\infty)} \cdot \mathbf{R}, \quad (5)$$

$$P \rightarrow P^{(\infty)}, \quad (6)$$

as $\mathbf{R} \rightarrow \infty$. We proceed now to nondimensionalize the above equations, as follows. The only characteristic length of the problem is the rest drop radius r_0 , which is then chosen as the unit length. Stresses are scaled with the interfacial stress σ/r_0 . External and internal velocities are scaled *differently*, by using σ/η , and $\sigma/\hat{\eta}$, respectively.

With these choices, the above equations stay unaltered (though the constitutive parameters η , $\hat{\eta}$, and σ disappear), except for Eq. (2) at the interface and Eq. (5) at infinity, which become, respectively,

$$\mathbf{V} = \frac{1}{\lambda} \hat{\mathbf{V}}, \quad (2')$$

$$\mathbf{V} \rightarrow \frac{r_0 \eta |\mathbf{L}_{\text{DIM}}^{(\infty)}|}{\sigma} \mathbf{L}^{(\infty)} \cdot \mathbf{R}, \quad (5')$$

where $\lambda = \hat{\eta}/\eta$ is the viscosity ratio, and $|\mathbf{L}_{\text{DIM}}^{(\infty)}|$ is the *dimensional* magnitude (or “strength”) of the imposed flow at infinity, a constant in steady-state situations as considered here (see later for possible changes of frame). The nondimensional group appearing in Eq. (5') is the well-known capillary number $Ca \equiv r_0 \eta |\mathbf{L}_{\text{DIM}}^{(\infty)}|/\sigma$, ratio of viscous to interfacial stresses.

It should be noted that the nondimensionalization chosen in this paper is different from the one more commonly adopted. Many authors^{10–13} take $\eta |\mathbf{L}_{\text{DIM}}^{(\infty)}|$ as the unit of stress. The reason why we prefer to use the constitutive quantity σ/r_0 to scale the stresses is simply that in such a way we automatically maintain the same nondimensionalization both at equilibrium and during flow. This is not the case with the alternative choice that introduces in the boundary condition of Eq. (4) a Ca^{-1} factor in the LHS of the equation (see Ref. 16), which is very disturbing for expansion purposes.

III. THE EXPANSION PROCEDURE

We proceed with the usual perturbation technique in order to solve the fluid dynamics problem in the so-called “weak flow limit,” with $Ca \ll 1$ as the *expansion parameter*, and $\lambda = O(1)$. The condition of small Ca implies small drop deformations from the unperturbed spherical shape, because interfacial forces are dominant. (Small deformations of the drop are also obtained under the condition $Ca = O(1)$ and $\lambda \rightarrow \infty$, i.e., for highly viscous drops.^{7,14,17} This case will not be considered in the following.) Up to second order in Ca , we write (trivial expansion equations are here reported mostly in order to define coefficients)

$$P = P_{\text{eq}} + Ca P_T + Ca^2 p, \quad (7)$$

$$\mathbf{V} = Ca \mathbf{V}_T + Ca^2 \mathbf{v}, \quad (8)$$

and analogous expansions [which will be referred to as Eqs. (7') and (8')] for the inner fields. (The subindex T of the coefficients of the first order problem stands for Taylor.) As regards the interface between the two fluids, it is convenient to describe it in a set of axes that moves with the center of the drop, and to choose a spherical coordinate representation. The position vector \mathbf{r} is written as $r\mathbf{u}$, with \mathbf{u} a unit vector. The drop surface is then given by $F = r - (1 + \Delta) = 0$, with Δ the “deviation” from the rest sphere. We can now rewrite the interface equation $F(\mathbf{r}, t) = 0$ in the form

$$r \equiv 1 + \Delta = 1 + Ca F_T + Ca^2 f. \quad (9)$$

Expansions analogous to that in Eq. (9) (in fact, deduced from it) will also hold for the surface-related quantities \mathbf{N} and $\mathbf{N} \nabla \cdot \mathbf{N}$ which appear in the interfacial boundary conditions; see Eqs. (3) and (4). We write

$$\mathbf{N} = \mathbf{u} + Ca \mathbf{N}_T + Ca^2 \mathbf{n}, \quad (10)$$

$$\mathbf{N}\nabla\cdot\mathbf{N}=2\mathbf{u}+Ca\mathbf{G}_T+Ca^2\mathbf{g}. \quad (11)$$

Notice that all the expansion coefficients of the interfacial quantities (i.e., F_T , f , \mathbf{N}_T , \mathbf{n} , \mathbf{G}_T , \mathbf{g}) only depend on the localization on the interface, i.e., on the \mathbf{u} vector, whereas the coefficients in Eqs. (7) and (8) [and those in Eqs. (7) and (8)] depend on the position vector $\mathbf{R}=\mathbf{R}\mathbf{u}$. Needless to say, all such quantities parametrically depend on the viscosity ratio λ , and in the general case also on time.

The expansions in Eqs. (7)–(11) are now inserted in the balance equations and boundary conditions given in the preceding section. They generate a hierarchy of problems at the zeroth order (statics: Laplace formula), first order (Taylor solution⁷), and second order in the capillary number Ca . As in all perturbative calculations, these problems have to be solved *sequentially*: the first order problem in Ca involves the equilibrium solution, and the second order problem involves Taylor solution [see Eqs. (14)–(19)].

As far as balance equations are concerned, substitution of the expansions given in Eqs. (7) and (8) (and the corresponding ones for the inner fluid) into the nondimensional version of Eqs. (1) [and Eq. (1)] is trivial. Beyond Laplace equilibrium formula, the resulting balance equations in flow situations are identical at order Ca and Ca^2 , and identical to Eqs. (1) and (1). As a matter of fact, balance equations maintain the same form at whatever order in Ca .

Concerning the boundary conditions, we immediately see that Eqs. (5) and (6) dictate the following conditions at infinity:

$$\mathbf{V}_T \rightarrow \mathbf{V}^{(\infty)}, \quad P_T \rightarrow P^{(\infty)} \quad \text{at order } Ca \quad (12)$$

and

$$\mathbf{v} \rightarrow \mathbf{0}, \quad p \rightarrow 0 \quad \text{at order } Ca^2. \quad (13)$$

On the other hand, some attention must be paid to write down the boundary conditions at order Ca and Ca^2 at the interface. The mere substitution of the expansions given by Eqs. (7), (8), (10), and (11) into the nondimensional version of Eqs. (2)–(4) is not the end of the story. Indeed, since all fields in Eqs. (2)–(4) must be evaluated at the actual interface $r(\mathbf{u}, t)$, and since $r(\mathbf{u}, t)$ itself is given by an expansion in Ca [see Eq. (9)], all fields in Eqs. (2)–(4) must themselves be expanded around the equilibrium value $r=1$, at any \mathbf{u} . Once these last expansions are performed, we obtain boundary conditions at the spherical surface $r=1$ that are *asymptotically equivalent* to the exact boundary conditions at the deformed surface $r(\mathbf{u}, t)$. When this just described “domain perturbation” technique^{6,16} is applied, we obtain

$$\mathbf{V}_T = \frac{1}{\lambda} \hat{\mathbf{V}}_T, \quad (14)$$

$$\mathbf{V}_T \cdot \mathbf{u} = \left(\frac{1}{|\nabla F|} \frac{\partial F}{\partial t} \right)_{\text{first order}}, \quad (15)$$

$$\mathbf{G}_T = (\hat{P}_{\text{eq}} - P_{\text{eq}}) \mathbf{N}_T + (\hat{P}_T - P_T) \mathbf{u} + 2(\mathbf{D}_T - \hat{\mathbf{D}}_T) \cdot \mathbf{u} \quad (16)$$

at order Ca (with \mathbf{D}_T and $\hat{\mathbf{D}}_T$ calculated from Taylor velocities), and

$$\mathbf{v} + F_T \frac{\partial \mathbf{V}_T}{\partial r} = \frac{1}{\lambda} \left(\hat{\mathbf{v}} + F_T \frac{\partial \hat{\mathbf{V}}_T}{\partial r} \right), \quad (17)$$

$$\mathbf{v} \cdot \mathbf{u} + \mathbf{V}_T \cdot \mathbf{N}_T + F_T \frac{\partial \mathbf{V}_T}{\partial r} \cdot \mathbf{u} = \left(\frac{1}{|\nabla F|} \frac{\partial F}{\partial t} \right)_{\text{second order}}, \quad (18)$$

$$\begin{aligned} \mathbf{g} = & (\hat{P}_{\text{eq}} - P_{\text{eq}}) \mathbf{n} + (\hat{P}_T - P_T) \mathbf{N}_T + \left[(\hat{p} - p) \right. \\ & \left. + F_T \frac{\partial (\hat{P}_T - P_T)}{\partial r} \right] \mathbf{u} + 2(\mathbf{D}_T - \hat{\mathbf{D}}_T) \cdot \mathbf{N}_T + 2 \left[(\mathbf{d} - \hat{\mathbf{d}}) \right. \\ & \left. + F_T \frac{\partial (\mathbf{D}_T - \hat{\mathbf{D}}_T)}{\partial r} \right] \cdot \mathbf{u} \end{aligned} \quad (19)$$

at order Ca^2 (with \mathbf{d} and $\hat{\mathbf{d}}$ calculated from the velocities \mathbf{v} and $\hat{\mathbf{v}}$, respectively), where all Eqs. (14)–(19) must be evaluated at the spherical (virtual) interface $r=1$.

Notice that, in the RHS of both Eqs. (15) and (18), we have not reported any explicit expression for the quantity $(\partial F / \partial t) / |\nabla F|$ at first and second order, respectively. To write such terms in time-dependent situations, additional assumptions would be required (see, e.g., Refs. 11 and 17). Since we are interested in steady-state calculations only, such terms will here be set to zero.

IV. PRESSURE AND VELOCITY FIELDS

The solution to the problem outlined above will be found in the following way. First, as is classically done,¹⁶ we reduce the Navier–Stokes equations to a Laplace equation for the pressure plus a Poisson equation for the velocity, to be solved sequentially (see Sec. VI). Next, however, we proceed in the calculations by an approach different from (and simpler of) the classical cumbersome expansion in spherical harmonics.

Since we are only concerned with steady-state situations (in some appropriate frame of reference), pressure and velocity fields must be completely determined by the “driving forces” at infinity, i.e., by the imposed *constant* velocity gradient $\mathbf{L}^{(\infty)}$ (the constant pressure $P^{(\infty)}$ can be taken to be zero with no loss of generality). On the other hand, we can easily write down (see below) the tensorial representations of these fields, up to second order, in terms of *time-dependent* $\mathbf{L}^{(\infty)}(t)$ and $\dot{\mathbf{L}}^{(\infty)}(t)$ when an arbitrary rotation (or change of frame) is superposed on the steady flow. Because of rotational invariance requirements,¹⁵ instead of tensors $\mathbf{L}^{(\infty)}(t)$ and $\dot{\mathbf{L}}^{(\infty)}(t)$, tensors $\mathbf{D}^{(\infty)}(t)$ and $\dot{\mathbf{D}}^{(\infty)}(t)$, and the vorticity tensor $\mathbf{W}^{(\infty)}(t)$ will in fact appear. (All tensors “at infinity” appearing in the equations hereafter will in general depend on time. For brevity of notations, however, we will not indicate this t dependency.)

Let us consecutively consider the Ca and Ca^2 cases. At first order in Ca , rotational invariance gives

$$P_T(R, \mathbf{u}) = \Pi(R) \mathbf{D}^{(\infty)} : \mathbf{u}\mathbf{u}, \quad (20)$$

$$\mathbf{V}_T(R, \mathbf{u}) = V_1(R) \mathbf{D}^{(\infty)} : \mathbf{u}\mathbf{u}\mathbf{u} + V_2(R) \mathbf{D}^{(\infty)} \cdot \mathbf{u} + R \mathbf{W}^{(\infty)} \cdot \mathbf{u}, \quad (21)$$

and

$$\hat{P}_T(\mathbf{R}, \mathbf{u}) = \hat{\Pi}(R) \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u}, \quad (22)$$

$$\hat{\mathbf{V}}_T(R, \mathbf{u}) = \hat{\mathbf{V}}_1(R) \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} \mathbf{u} + \hat{\mathbf{V}}_2(R) \mathbf{D}^{(\infty)} \cdot \mathbf{u} + \lambda R \mathbf{W}^{(\infty)} \cdot \mathbf{u} \quad (23)$$

for the outer and inner fields, respectively, where all the radial functions remain to be determined through actual solution of the equations of motion. It is worth noting that the rightmost terms in Eqs. (21) and (23) account for (arbitrary) rotation of the velocity field at infinity. For future reference, it is also noticed that, at first order, no \mathbf{u} -independent pressure term builds up due to motion. Indeed, possible scalar terms proportional to $\text{Tr}(\mathbf{D}^{(\infty)})$ [$\text{Tr}(\cdot \cdot)$ is the trace operator] vanish because of the incompressibility condition.

In going up to second order in Ca , it is worth emphasizing that, since rigid rotations are already accounted for at first order, $\mathbf{W}^{(\infty)}$ can only appear *inside* the second Rivlin–Ericksen tensor $\mathbf{A}_2^{(\infty)}$, which is defined as¹⁸

$$\mathbf{A}_2^{(\infty)} = 2(\dot{\mathbf{D}}^{(\infty)} - \mathbf{W}^{(\infty)} \cdot \mathbf{D}^{(\infty)} + \mathbf{D}^{(\infty)} \cdot \mathbf{W}^{(\infty)}) + 4\mathbf{D}^{(\infty)} \cdot \mathbf{D}^{(\infty)}. \quad (24)$$

Although the product $\mathbf{D}^{(\infty)} \cdot \mathbf{D}^{(\infty)}$ appears in the definition of $\mathbf{A}_2^{(\infty)}$, the general second order expansion must also include an independent $\mathbf{D}^{(\infty)} \cdot \mathbf{D}^{(\infty)}$ tensor. We thus obtain

$$p(\mathbf{R}, \mathbf{u}) = p_1(R) \mathbf{D}^{(\infty)} \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} \mathbf{u} \mathbf{u} + p_2(R) \mathbf{D}^{(\infty)} \cdot \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} + p_3(R) \mathbf{D}^{(\infty)} : \mathbf{D}^{(\infty)} + p_4(R) \mathbf{A}_2^{(\infty)} : \mathbf{u} \mathbf{u}, \quad (25)$$

$$\begin{aligned} \mathbf{v}(R, \mathbf{u}) = & v_1(R) \mathbf{D}^{(\infty)} \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} \mathbf{u} \mathbf{u} + v_2(R) \mathbf{D}^{(\infty)} \cdot \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} \mathbf{u} + v_3(R) \mathbf{D}^{(\infty)} : \mathbf{D}^{(\infty)} \mathbf{u} \\ & + v_4(R) \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} \mathbf{D}^{(\infty)} \cdot \mathbf{u} + v_5(R) \mathbf{D}^{(\infty)} \cdot \mathbf{D}^{(\infty)} \cdot \mathbf{u} \\ & + v_6(R) \mathbf{A}_2^{(\infty)} : \mathbf{u} \mathbf{u} \mathbf{u} + v_7(R) \mathbf{A}_2^{(\infty)} \cdot \mathbf{u} \end{aligned} \quad (26)$$

and analogous expressions, with $\hat{p}_i(R)$ and $\hat{v}_i(R)$, for the inner fields. Notice that the terms containing $p_3(R)$ and $\hat{p}_3(R)$ constitute \mathbf{u} -independent pressure terms that arise at order Ca^2 . This point will be taken up again in Sec. VI.

V. DROP SHAPE AND DROP VOLUME CONSERVATION

Before proceeding in the solution for the second order fields p and \mathbf{v} (and \hat{p} , $\hat{\mathbf{v}}$) considered in the preceding section, we need to expand to the same order the drop shape. We also need to account for drop volume conservation. Since the drop shape is described by a scalar function $r(\mathbf{u})$ [see Eq. (9)], the most general rotationally invariant expansion of $r(\mathbf{u})$ has the same tensorial form as that for $P(\mathbf{u})$ previously considered [Eqs. (7), (20), and (25)]. Hence

$$\begin{aligned} r(\mathbf{u}) = & 1 + Ca T \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} + Ca^2 (s_1 \mathbf{D}^{(\infty)} \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} \mathbf{u} \mathbf{u} \\ & + s_2 \mathbf{D}^{(\infty)} \cdot \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} + s_3 \mathbf{D}^{(\infty)} : \mathbf{D}^{(\infty)} + s_4 \mathbf{A}_2^{(\infty)} : \mathbf{u} \mathbf{u}), \end{aligned} \quad (27)$$

where the five scalar coefficients, T plus the four s_i , only depend on the viscosity ratio λ .

Let us now explicitly calculate the constraint imposed on these coefficients by drop volume conservation, i.e., by the condition

$$\int d\mathbf{u} \int_0^{r(\mathbf{u})} dr r^2(\mathbf{u}) = \frac{4}{3} \pi, \quad (28)$$

where the integral over \mathbf{u} is a shorthand notation for the double integral over the spherical solid angle Ω ($d\mathbf{u} = d\Omega = \sin \theta d\theta d\varphi$). By performing the integration in Eq. (28) with the expansion of $r(\mathbf{u})$ in Eq. (27) we find that, at first order, volume conservation does not introduce constraints (a result known since the work of Taylor⁷), while at second order the following relationship among the drop shape coefficients must be satisfied:

$$2(T^2 + s_1) + 5s_2 + 15s_3 + 20s_4 = 0. \quad (29)$$

Equation (29) is derived in Appendix A.

VI. THE COMPLETE SECOND-ORDER SOLUTION

In Secs. IV and V we have shown that the angular dependencies of pressure and velocity fields and the generic drop shape are known beforehand, from general invariance arguments. Therefore, we are left with the problem of determining scalar radial functions only, plus the scalar coefficients in Eq. (27) for the drop shape [subjected to the constraint of Eq. (29)]. For the reader convenience, we report in Appendix B the Taylor solution⁷ to the first order problem, expressed in the form used in this paper. We outline here the calculation procedure at order Ca^2 , and give the complete solution.

To start with, the radial dependencies of all 22 radial functions p_i and v_i , and \hat{p}_i and \hat{v}_i (for the outer and inner fields, respectively) have to be determined. To this aim, we first insert the general expressions of the second-order pressure fields into the Laplace equations mentioned above (see the beginning of Sec. IV), e.g., $\nabla^2 p = 0$ for the external pressure. After some computations [see Eq. (C2) for the Laplacians in Appendix C], the tensorial representation of the scalar quantity $\nabla^2 p$ is obtained, which is then equated term by term to zero. A set of second-order ordinary differential equations for the functions p_i and \hat{p}_i is thus obtained [Eq. (D1) in Appendix D], and easily solved. By further imposing that the external pressure goes to zero at infinity [see Eq. (13)], and that the internal pressure is finite at $R=0$, we find

$$\begin{aligned} p_1(R) &= s_{p1} R^{-5}, \\ p_2(R) &= s_{p2} R^{-3} - (4/7) s_{p1} R^{-5}, \\ p_3(R) &= s_{p3} R^{-1} - (1/3)(s_{p2} + 4s_{p4}) R^{-3} + (2/35) s_{p1} R^{-5}, \\ p_4(R) &= s_{p4} R^{-3} \end{aligned} \quad (30)$$

and

$$\begin{aligned}
\hat{p}_1(R) &= \hat{s}_{p1} R^4, \\
\hat{p}_2(R) &= \hat{s}_{p2} R^2 - (4/7) \hat{s}_{p1} R^4, \\
\hat{p}_3(R) &= \hat{s}_{p3} - (1/3)(\hat{s}_{p2} + 4\hat{s}_{p4}) R^2 + (2/35) \hat{s}_{p1} R^4, \\
\hat{p}_4(R) &= \hat{s}_{p4} R^2,
\end{aligned} \quad (31)$$

where the scalar coefficients s_{pi} and \hat{s}_{pi} only depend on λ .

By using Eqs. (30) and (31), we now calculate ∇p and $\nabla \hat{p}$ [see Eq. (C1) in Appendix C], to be inserted as “density terms” into the vectorial Poisson equations for the velocity. By also using Eq. (26) for \mathbf{v} , and the equivalent one for $\hat{\mathbf{v}}$, the tensorial form of the Poisson equations is computed. Equating coefficients of like tensorial terms in such equations gives a set of second-order ordinary differential equations [see Eq. (D2) in Appendix D], from which the functions v_i and \hat{v}_i are calculated. Under the conditions that the external velocity goes to zero at infinity [see Eq. (13)], and that the internal velocity is finite at $R=0$, we obtain

$$\begin{aligned}
v_1(R) &= s_{v1} R^{-6} + (1/2) s_{p1} R^{-4}, \\
v_2(R) &= s_{v2} R^{-4} + (1/2) s_{p2} R^{-2} - (4/9) s_{v1} R^{-6}, \\
v_3(R) &= s_{v3} R^{-2} - (1/35)(s_{p1} + 7s_{v2} + 28s_{v6}) R^{-4} \\
&\quad + (2/63) s_{v1} R^{-6}, \\
v_4(R) &= s_{v4} R^{-4} - (4/9) s_{v1} R^{-6}, \\
v_5(R) &= s_{v5} R^{-2} - (2/35)(2s_{p1} + 7s_{v2} + 7s_{v4}) R^{-4} \\
&\quad + (8/63) s_{v1} R^{-6}, \\
v_6(R) &= s_{v6} R^{-4} + (1/2) s_{p4} R^{-2}, \\
v_7(R) &= s_{v7} R^{-2} - (2/5) s_{v6} R^{-4}
\end{aligned} \quad (32)$$

and

$$\begin{aligned}
\hat{v}_1(R) &= \hat{s}_{v1} R^5, \\
\hat{v}_2(R) &= \hat{s}_{v2} R^3 - (4/63)(\hat{s}_{p1} + 7\hat{s}_{v1}) R^5, \\
\hat{v}_3(R) &= \hat{s}_{v3} R - (1/15)(\hat{s}_{p2} + 4\hat{s}_{p4} + 3\hat{s}_{v2} + 12\hat{s}_{v6}) R^3 \\
&\quad + (2/315)(2\hat{s}_{p1} + 5\hat{s}_{v1}) R^5, \\
\hat{v}_4(R) &= \hat{s}_{v4} R^3 + (2/9)(\hat{s}_{p1} - 2\hat{s}_{v1}) R^5, \\
\hat{v}_5(R) &= \hat{s}_{v5} R + (1/5)(\hat{s}_{p2} - 2\hat{s}_{v2} - 2\hat{s}_{v4}) R^3 + (4/63) \\
&\quad \times (-\hat{s}_{p1} + 2\hat{s}_{v1}) R^5, \\
\hat{v}_6(R) &= \hat{s}_{v6} R^3, \\
\hat{v}_7(R) &= \hat{s}_{v7} R + (1/5)(\hat{s}_{p4} - 2\hat{s}_{v6}) R^3,
\end{aligned} \quad (33)$$

where also the new scalar coefficients s_{vi} and \hat{s}_{vi} only depend on λ .

It should be noted here that the incompressibility condition for both the outer and inner fluids has not yet been imposed on the results of Eqs. (32) and (33). Such conditions, in terms of the v_i and \hat{v}_i functions, are written in Eq. (D3). Substitution of Eqs. (32) and (33) into Eq. (D3) leads to a set of algebraic relationships among (some of) the scalar coefficients s_{vi} and \hat{s}_{vi} , and s_{pi} and \hat{s}_{pi} .

To complete the solution of the drop problem, we must now determine all the scalar coefficients s_{pi} , \hat{s}_{pi} , s_{vi} , and \hat{s}_{vi} , appearing in Eqs. (30)–(33), plus the drop shape coefficients s_1, \dots, s_4 in Eq. (27), in terms of the viscosity ratio λ . This is achieved by imposing the interfacial boundary conditions, Eqs. (17)–(19), the constraint of drop volume conservation, Eq. (29), and the just mentioned algebraic relationships arising from the constraint of fluid incompressibility. [Appendix E provides the expressions of the “geometric” quantities \mathbf{n} and \mathbf{g} appearing in the interfacial boundary conditions; see Eqs. (10) and (11).]

The ensuing algebra, though straightforward, is still rather lengthy. We have chosen to perform many of the required manipulations by means of MATHEMATICA®. The final results are listed in Appendix F. It is worth noticing that, since the coefficient \hat{s}_{p3} comes out nonzero, a constant pressure term $\hat{s}_{p3} \mathbf{D}^{(\infty)} : \mathbf{D}^{(\infty)}$ is predicted to arise inside the drop in response to flow, at order Ca^2 .

VII. DISCUSSION AND CONCLUSIONS

We have presented here the complete solution (at order Ca^2) of the fluidodynamic problem of a single drop immersed in a flowing immiscible fluid, for the Newtonian case, under steady state. It was possible to present the perturbation procedure in sufficient detail, because we took full advantage of the simplification introduced by using rotational invariance. Indeed, the latter leads to a compact tensorial representation of the pressure and velocity fields, and of the drop shape, in terms of the imposed flow field “at infinity.”

The traditional spherical harmonics expansion method for the drop problem at order Ca^2 has a long and tricky history,^{9–13} as reviewed by Rallison.¹⁴ Inclusion of volume conservation by Frankel and Acrivos,¹¹ and of an incremental \mathbf{R} -independent inner pressure due to flow by Barthes-Biesel and Acrivos^{12,13} (see the comment on the \hat{s}_{p3} coefficient at the end of the last section), marked significant progress. Minor differences (numerically irrelevant) of our results with respect to those of Barthes-Biesel and Acrivos^{12,13} remain, but we are unable to locate with certainty the source of these discrepancies. It should be mentioned in this respect that one of the boundary conditions (the one on normal forces at the interface) is formulated differently in the two approaches, possibly as a consequence of the different nondimensionalization. How much such a difference may influence the overall solution remains however open to question. Concerning comparison with experiments,^{20,21} the numerical differences between our solution and that of Barthes-Biesel and Acrivos^{12,13} are so small that both can be considered in full agreement with data.

As mentioned repeatedly, the adoption in this paper of a tensorial representation for the unknown fields, and for the drop shape, greatly simplifies the perturbative calculations. We summarize here the main advantages: (i) the mathematical problem is reduced to the determination of scalar radial functions only, and of the scalar coefficients for the drop shape; (ii) the relevant equations to be solved are readily obtained by writing down the tensorial equations *term by*

term, and equating to zero the corresponding coefficients (see Appendix D). The latter procedure should be compared to that resulting from the spherical harmonics expansion (and Lamb's general solution⁸), where angular integrations must be performed to exploit the orthogonality properties of spherical harmonics. The greater simplicity of the method developed here makes us reasonably confident of the correctness of our algebra as compared to previous results.

One disadvantage of our method however exists. Indeed, for unsteady flows which evolve "on a surface tension time scale" $\tau = r_0 \eta / \sigma$, pressure and velocity fields become dependent on the *history* of the velocity gradient at infinity, and the usefulness of the invariance method becomes dubious. Solution of the drop problem in transient flows through the classical spherical harmonics expansion can then perhaps remain more convenient.

As mentioned in the Introduction, even with Newtonian component fluids, the overall system, made up of many non-interacting drops, is a non-Newtonian fluid, because of the elasticity arising from the interface. Of course, elasticity effects can independently arise from the *constitutive* behavior of the components, i.e., when non-Newtonian fluids are considered. The drop problem has never been solved for such a case. This is explained by noting that the analogous of Lamb's general solution⁸ does not exist for non-Newtonian fluids. The systematic mathematical approach presented here, however, is readily extendable to the slow-flow case with non-Newtonian components. Some results of these new calculations have already been presented,²² and the complete solution will be given in a forthcoming paper.

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APPENDIX A: DROP VOLUME CONSERVATION

The inner integral in Eq. (28) of the text gives $r^3(\mathbf{u})/3$, which up to second order becomes [see Eq. (27)]

$$\begin{aligned} \frac{r^3(\mathbf{u})}{3} &= \frac{1}{3} + Ca T \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} \\ &+ Ca^2 [(T^2 + s_1) \mathbf{D}^{(\infty)} \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} \mathbf{u} \mathbf{u} \\ &+ s_2 \mathbf{D}^{(\infty)} \cdot \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} + s_3 \mathbf{D}^{(\infty)} : \mathbf{D}^{(\infty)} + s_4 \mathbf{A}_2^{(\infty)} : \mathbf{u} \mathbf{u}]. \end{aligned} \quad (\text{A1})$$

Since $\int d\mathbf{u} = 4\pi$, the zeroth order term of Eq. (A1) (i.e., $1/3$) already satisfies Eq. (28). Hence, the following scalar conditions must hold true:

$$\mathbf{D}^{(\infty)} : \int d\mathbf{u} \mathbf{u} \mathbf{u} = 0, \quad (\text{A2})$$

$$\begin{aligned} (T^2 + s_1) \mathbf{D}^{(\infty)} \mathbf{D}^{(\infty)} : \int d\mathbf{u} \mathbf{u} \mathbf{u} \mathbf{u} \mathbf{u} \\ + (s_2 \mathbf{D}^{(\infty)} \cdot \mathbf{D}^{(\infty)} + s_4 \mathbf{A}_2^{(\infty)}) : \int d\mathbf{u} \mathbf{u} \mathbf{u} \\ + s_3 \mathbf{D}^{(\infty)} : \mathbf{D}^{(\infty)} \int d\mathbf{u} = 0. \end{aligned} \quad (\text{A3})$$

To proceed, the following identities are needed:¹⁹

$$\int d\mathbf{u} u_i u_j = \frac{4}{3} \pi \delta_{ij}, \quad (\text{A4})$$

$$\int d\mathbf{u} u_i u_j u_k u_l = \frac{4}{15} \pi (\delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}). \quad (\text{A5})$$

It is then immediately verified that Eq. (A2) is automatically satisfied, because of the incompressibility condition $\text{Tr}(\mathbf{D}^{(\infty)}) = 0$. Thus, drop volume conservation is automatic at first order in Ca . Conversely, by inserting Eqs. (A4) and (A5) in Eq. (A3), and using the relationship $\text{Tr}(\mathbf{A}_2^{(\infty)}) = 4 \mathbf{D}^{(\infty)} : \mathbf{D}^{(\infty)}$ [see Eq. (24)], we find

$$[2(T^2 + s_1) + 5s_2 + 15s_3 + 20s_4] \mathbf{D}^{(\infty)} : \mathbf{D}^{(\infty)} = 0. \quad (\text{A6})$$

Equation (A6) must be valid for any imposed velocity field at infinity, i.e., for arbitrary $\mathbf{D}^{(\infty)} : \mathbf{D}^{(\infty)}$, hence the term in square brackets must be zero, which constitutes Eq. (29) of the text.

APPENDIX B: TAYLOR (1932) FIRST-ORDER SOLUTION

The first-order solution by Taylor, expressed in the tensorial form adopted in the paper, is as follows.

(i) Outer fields

$$P_T(R, \mathbf{u}) = T_P \frac{1}{R^3} \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u}, \quad (\text{B1})$$

$$\begin{aligned} \mathbf{V}_T(R, \mathbf{u}) &= \left(T_1 \frac{1}{R^4} + \frac{T_P}{2} \frac{1}{R^2} \right) \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} \mathbf{u} \\ &+ \left(-\frac{2T_1}{5} \frac{1}{R^4} + R \right) \mathbf{D}^{(\infty)} \cdot \mathbf{u} + R \mathbf{W}^{(\infty)} \cdot \mathbf{u}. \end{aligned} \quad (\text{B2})$$

(ii) Inner fields

$$\hat{P}_T(R, \mathbf{u}) = \hat{T}_P R^2 \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u}, \quad (\text{B3})$$

$$\begin{aligned} \hat{\mathbf{V}}_T(R, \mathbf{u}) &= \hat{T}_1 R^3 \mathbf{D}^{(\infty)} : \mathbf{u} \mathbf{u} \mathbf{u} + \left(\hat{T}_2 R + \frac{\hat{T}_P - 2\hat{T}_1}{5} R^3 \right) \mathbf{D}^{(\infty)} \\ &\cdot \mathbf{u} + \lambda R \mathbf{W}^{(\infty)} \cdot \mathbf{u}. \end{aligned} \quad (\text{B4})$$

(iii) Field coefficients

$$\begin{aligned} T_P &= -\frac{2+5\lambda}{1+\lambda}, \quad T_1 = \frac{5\lambda}{2(1+\lambda)}, \\ \hat{T}_P &= \frac{21\lambda}{2(1+\lambda)}, \quad \hat{T}_1 = -\frac{\lambda}{1+\lambda}, \quad \hat{T}_2 = -\frac{3\lambda}{2(1+\lambda)}. \end{aligned} \quad (\text{B5})$$

(iv) Drop shape

$$r(\mathbf{u}) = 1 + Ca \frac{16 + 19\lambda}{8(1 + \lambda)} \mathbf{D}^{(\infty)} : \mathbf{u}\mathbf{u}. \quad (\text{B6})$$

APPENDIX C: USEFUL FORMULAS FOR THE GRADIENT AND LAPLACIAN OF POLYADIC FUNCTIONS

In the computations of the paper, gradients and Laplacians of polyadic functions $f(R)\mathbf{u} \cdots \mathbf{u}$ [with $f(R)$ a generic scalar radial function] repeatedly appear. We give in this appendix some of these derivatives. Indicinal notation is adopted for clarity. The gradient operator will be denoted by d_α , and the Laplacian operator by L .

(i) Gradients

$$\begin{aligned} d_\alpha(fu_i) &= \left(\frac{df}{dR} - \frac{f}{R} \right) u_i u_\alpha + \frac{f}{R} \delta_{i\alpha}, \\ d_\alpha(fu_i u_j) &= \left(\frac{df}{dR} - \frac{2f}{R} \right) u_i u_j u_\alpha + \frac{f}{R} (u_i \delta_{j\alpha} + u_j \delta_{i\alpha}), \\ d_\alpha(fu_i u_j u_k) &= \left(\frac{df}{dR} - \frac{3f}{R} \right) u_i u_j u_k u_\alpha + \frac{f}{R} (u_i u_j \delta_{k\alpha} \\ &\quad + u_k u_i \delta_{j\alpha} + u_j u_k \delta_{i\alpha}) \dots \end{aligned} \quad (\text{C1})$$

Formulas for higher order polyadic are obtained by induction: the polyadic order p appears as a numerical coefficient in the first brackets; the second brackets contains p deltas $\delta_{(\cdot)\alpha}$.

(ii) Laplacians

$$\begin{aligned} L(fu_i) &= \left(\frac{d^2 f}{dR^2} + \frac{2}{R} \frac{df}{dR} - \frac{2f}{R^2} \right) u_i, \\ L(fu_i u_j) &= \left(\frac{d^2 f}{dR^2} + \frac{2}{R} \frac{df}{dR} - \frac{6f}{R^2} \right) u_i u_j + \frac{2f}{R^2} \delta_{ij}, \\ L(fu_i u_j u_k) &= \left(\frac{d^2 f}{dR^2} + \frac{2}{R} \frac{df}{dR} - \frac{12f}{R^2} \right) u_i u_j u_k + \frac{2f}{R^2} (u_i \delta_{jk} \\ &\quad + u_k \delta_{ij} + u_j \delta_{ik}) \dots \end{aligned} \quad (\text{C2})$$

Here again, formulas for higher-order polyadic are obtained by induction: the product $p(p+1)$ appears as the last numerical coefficient in the first brackets; the second brackets contain all possible deltas.

APPENDIX D: DIFFERENTIAL EQUATIONS FOR PRESSURE AND VELOCITY FIELDS: THE INCOMPRESSIBILITY CONSTRAINT

By inserting the general second-order tensorial expressions for pressure and velocity fields into the balance equations, and equating coefficients of like tensorial terms, a set of scalar ordinary differential equations is obtained, which is reported in this appendix.

(i) From the equation $\nabla^2 p = 0$ (order Ca^2):

$$\frac{d^2 p_1}{dR^2} + \frac{2}{R} \frac{dp_1}{dR} - \frac{20p_1}{R^2} = 0,$$

$$\begin{aligned} \frac{d^2 p_2}{dR^2} + \frac{2}{R} \frac{dp_2}{dR} - \frac{6p_2}{R^2} + \frac{8p_1}{R^2} &= 0, \\ \frac{d^2 p_3}{dR^2} + \frac{2}{R} \frac{dp_3}{dR} + \frac{2p_2 + 8p_4}{R^2} &= 0, \\ \frac{d^2 p_4}{dR^2} + \frac{2}{R} \frac{dp_4}{dR} - \frac{6p_4}{R^2} &= 0. \end{aligned} \quad (\text{D1})$$

(The same equations also hold for the inner fields $\hat{p}_1, \dots, \hat{p}_4$, i.e., from the equation $\nabla^2 \hat{p} = 0$.)

(ii) From the equation $\nabla^2 \mathbf{v} = \nabla p$ (order Ca^2):

$$\begin{aligned} \frac{d^2 v_1}{dR^2} + \frac{2}{R} \frac{dv_1}{dR} - \frac{30v_1}{R^2} &= \frac{dp_1}{dR} - \frac{4p_1}{R}, \\ \frac{d^2 v_2}{dR^2} + \frac{2}{R} \frac{dv_2}{dR} - \frac{12v_2}{R^2} + \frac{8v_1}{R^2} &= \frac{dp_2}{dR} - \frac{2p_2}{R}, \\ \frac{d^2 v_3}{dR^2} + \frac{2}{R} \frac{dv_3}{dR} - \frac{2v_3}{R^2} + \frac{2v_2 + 8v_6}{R^2} &= \frac{dp_3}{dR}, \\ \frac{d^2 v_4}{dR^2} + \frac{2}{R} \frac{dv_4}{dR} - \frac{12v_4}{R^2} + \frac{8v_1}{R^2} &= \frac{4p_1}{R}, \\ \frac{d^2 v_5}{dR^2} + \frac{2}{R} \frac{dv_5}{dR} - \frac{2v_5}{R^2} + \frac{4v_2 + 4v_4}{R^2} &= \frac{2p_2}{R}, \\ \frac{d^2 v_6}{dR^2} + \frac{2}{R} \frac{dv_6}{dR} - \frac{12v_6}{R^2} &= \frac{dp_4}{dR} - \frac{2p_4}{R}, \\ \frac{d^2 v_7}{dR^2} + \frac{2}{R} \frac{dv_7}{dR} - \frac{2v_7}{R^2} + \frac{4v_6}{R^2} &= \frac{2p_4}{R}. \end{aligned} \quad (\text{D2})$$

(The same equations also hold for the inner fields $\hat{v}_1, \dots, \hat{v}_7$, i.e., from the equation $\nabla^2 \hat{\mathbf{v}} = \nabla \hat{p}$.) Solution of Eqs. (D1) and (D2) gives Eqs. (30)–(33) in the text.

(iii) From the equation $\nabla \cdot \mathbf{v} = 0$ (order Ca^2):

$$\begin{aligned} \frac{dv_1}{dR} + \frac{dv_4}{dR} + \frac{2v_1 - 3v_4}{R} &= 0, \\ \frac{dv_2}{dR} + \frac{dv_5}{dR} + \frac{2v_2 + 2v_4 - v_5}{R} &= 0, \\ \frac{dv_3}{dR} + \frac{2v_3 + v_5 + 4v_7}{R} &= 0 \\ \frac{dv_6}{dR} + \frac{dv_7}{dR} + \frac{2v_6 - v_7}{R} &= 0. \end{aligned} \quad (\text{D3})$$

(The same equations also hold for the inner fields $\hat{v}_1, \dots, \hat{v}_7$, i.e., from the equation $\nabla \cdot \hat{\mathbf{v}} = 0$.) The incompressibility as expressed here imposes constraints on the fields $v_i(R)$ and $\hat{v}_i(R)$, which are explicitly calculated by inserting Eqs. (32) and (33) in the text into Eq. (D3).

APPENDIX E: THE GEOMETRIC QUANTITIES \mathbf{n} AND \mathbf{g}

To evaluate the second-order boundary conditions at the interface, Eqs. (17)–(19), the second-order “geometric” quantities \mathbf{n} and \mathbf{g} must be calculated, whose definition is given in Eqs. (10) and (11) in the text. Calculation of these

quantities is slightly involved, because the “domain perturbation” technique⁶ has to be applied. We give here the resulting final expressions:

$$\begin{aligned} \mathbf{n}(\mathbf{u}) = & 4s_1 \mathbf{D}^{(\infty)} \mathbf{D}^{(\infty)} :: \mathbf{uuuuu} + (2s_2 - 2T^2) \mathbf{D}^{(\infty)} \\ & \cdot \mathbf{D}^{(\infty)} : \mathbf{uuu} + (-4s_1 + 2T^2) \mathbf{D}^{(\infty)} : \mathbf{uu} \mathbf{D}^{(\infty)} \cdot \mathbf{u} \\ & + (-2s_2) \mathbf{D}^{(\infty)} \cdot \mathbf{D}^{(\infty)} \cdot \mathbf{u} + 2s_4 \mathbf{A}_2^{(\infty)} : \mathbf{uuu} \\ & + (-2s_4) \mathbf{A}_2^{(\infty)} \cdot \mathbf{u}, \end{aligned} \quad (\text{E1})$$

$$\begin{aligned} \mathbf{g}(\mathbf{u}) = & (18s_1 - 2T^2) \mathbf{D}^{(\infty)} \mathbf{D}^{(\infty)} :: \mathbf{uuuuu} + (-8s_1 \\ & + 4s_2) \mathbf{D}^{(\infty)} \cdot \mathbf{D}^{(\infty)} : \mathbf{uuu} + (-2s_2 - 2s_3 \\ & - 8s_4) \mathbf{D}^{(\infty)} : \mathbf{D}^{(\infty)} \mathbf{u} + (-8T^2) \mathbf{D}^{(\infty)} : \mathbf{uu} \mathbf{D}^{(\infty)} \cdot \mathbf{u} \\ & + 4s_4 \mathbf{A}_2^{(\infty)} : \mathbf{uuu} + 2\mathbf{n}(\mathbf{u}). \end{aligned} \quad (\text{E2})$$

APPENDIX F: DEPENDENCE OF ALL SECOND-ORDER COEFFICIENTS ON THE VISCOSITY RATIO λ

Drop shape coefficients

$$\begin{aligned} s_1 = & \frac{16+19\lambda}{8(1+\lambda)} \frac{656+751\lambda}{216(1+\lambda)}, \\ s_2 = & \frac{16+19\lambda}{8(1+\lambda)} \frac{4712+\lambda(17\,967+\lambda(16\,360+3591\lambda))}{1890(1+\lambda)^2}, \\ s_3 = & -\frac{16+19\lambda}{8(1+\lambda)} \frac{2656+\lambda(8835+5855\lambda)}{3780(1+\lambda)^2}, \\ s_4 = & -\left(\frac{16+19\lambda}{8(1+\lambda)}\right)^2 \frac{3+2\lambda}{10}. \end{aligned} \quad (\text{F1})$$

Outer pressure coefficients

$$\begin{aligned} s_{p1} = & -\frac{16+19\lambda}{8(1+\lambda)} \frac{154+315\lambda}{9(1+\lambda)}, \\ s_{p2} = & -\frac{16+19\lambda}{8(1+\lambda)} \frac{31+\lambda(92+52\lambda)}{7(1+\lambda)^2}, \\ s_{p3} = & 0, \\ s_{p4} = & \left(\frac{16+19\lambda}{8(1+\lambda)}\right)^2 \frac{4}{5}. \end{aligned} \quad (\text{F2})$$

Inner pressure coefficients

$$\begin{aligned} \hat{s}_{p1} = & \frac{16+19\lambda}{8(1+\lambda)} \frac{385\lambda}{18(1+\lambda)}, \\ \hat{s}_{p2} = & \frac{16+19\lambda}{8(1+\lambda)} \frac{\lambda(51+\lambda(57+42\lambda))}{5(1+\lambda)^2}, \\ \hat{s}_{p3} = & -\frac{16+19\lambda}{8(1+\lambda)} \frac{-4+74\lambda}{15(1+\lambda)}, \\ \hat{s}_{p4} = & -\frac{16+19\lambda}{8(1+\lambda)} \frac{\lambda(63+42\lambda)}{20(1+\lambda)}. \end{aligned} \quad (\text{F3})$$

Outer velocity coefficients

$$\begin{aligned} s_{v1} = & \frac{16+19\lambda}{8(1+\lambda)} \frac{4+45\lambda}{2(1+\lambda)}, \\ s_{v2} = & \frac{16+19\lambda}{8(1+\lambda)} \frac{112+\lambda(466+318\lambda)}{21(1+\lambda)^2}, \\ s_{v3} = & \frac{16+19\lambda}{8(1+\lambda)} \frac{8+\lambda(82+50\lambda)}{70(1+\lambda)^2}, \\ s_{v4} = & \frac{16+19\lambda}{8(1+\lambda)} \frac{22+45\lambda}{9(1+\lambda)}, \\ s_{v5} = & 0, \\ s_{v6} = & -\frac{16+19\lambda}{8(1+\lambda)} \frac{2+3\lambda}{4(1+\lambda)}, \\ s_{v7} = & 0. \end{aligned} \quad (\text{F4})$$

Inner velocity coefficients

$$\begin{aligned} \hat{s}_{v1} = & -\frac{16+19\lambda}{8(1+\lambda)} \frac{14\lambda}{9(1+\lambda)}, \\ \hat{s}_{v2} = & -\frac{16+19\lambda}{8(1+\lambda)} \frac{\lambda(-494+\lambda(-458+252\lambda))}{315(1+\lambda)^2}, \\ \hat{s}_{v3} = & -\frac{16+19\lambda}{8(1+\lambda)} \frac{6\lambda(-1+\lambda)}{35(1+\lambda)^2}, \\ \hat{s}_{v4} = & -\frac{16+19\lambda}{8(1+\lambda)} \frac{80\lambda}{9(1+\lambda)}, \\ \hat{s}_{v5} = & -\frac{16+19\lambda}{8(1+\lambda)} \frac{\lambda(302+\lambda(454+224\lambda))}{70(1+\lambda)^2}, \\ \hat{s}_{v6} = & \frac{16+19\lambda}{8(1+\lambda)} \frac{\lambda(3+2\lambda)}{10(1+\lambda)}, \\ \hat{s}_{v7} = & \left(\frac{16+19\lambda}{8(1+\lambda)}\right)^2 \frac{2\lambda}{5}. \end{aligned} \quad (\text{F5})$$

In all of these coefficients, the factor $(16+19\lambda)/8(1+\lambda)$ entering Taylor first-order solution (see Appendix B) always appears. It was thought useful to show such feature explicitly.

¹W. R. Schowalter, *Mechanics of Non-Newtonian Fluids* (Pergamon, Oxford, 1978).

²J. M. Rallison, “The deformation of small viscous drops and bubbles in shear flows,” *Annu. Rev. Fluid Mech.* **16**, 45 (1984).

³H. A. Stone, “Dynamics of drop deformation and breakup in viscous fluids,” *Annu. Rev. Fluid Mech.* **26**, 65 (1994).

⁴M. R. Kennedy, C. Pozrikidis, and R. Skalak, “Motion and deformation of liquid drops, and the rheology of dilute emulsions in simple shear flow,” *Comput. Fluids* **23**, 251 (1994).

⁵W. S. J. Uijttewaal and E. J. Nijhof, “The motion of a droplet subjected to linear shear flow including the presence of a plane wall,” *J. Fluid Mech.* **302**, 45 (1995).

⁶D. D. Joseph, *Fluid Dynamics of Viscoelastic Liquids* (Springer-Verlag, New York, 1990).

⁷G. I. Taylor, “The viscosity of a fluid containing small drops of another fluid,” *Proc. R. Soc. London, Ser. A* **138**, 41 (1932).

⁸H. Lamb, *Hydrodynamics* (Dover, New York, 1945).

⁹C. E. Chaffey and H. Brenner, “A second-order theory for shear deformation of drops,” *J. Colloid Interface Sci.* **24**, 258 (1967).

- ¹⁰N. A. Frankel, Ph.D. dissertation, Stanford University, 1968.
- ¹¹N. A. Frankel and A. Acrivos, "The constitutive equation for a dilute emulsion," *J. Fluid Mech.* **44**, 65 (1970).
- ¹²D. Barthes-Biesel, Ph.D. dissertation, Stanford University, 1972.
- ¹³D. Barthes-Biesel and A. Acrivos, "Deformation and burst of a liquid droplet freely suspended in a linear shear field," *J. Fluid Mech.* **61**, 1 (1973).
- ¹⁴J. M. Rallison, "Note on the time-dependent deformation of a viscous drop which is almost spherical," *J. Fluid Mech.* **98**, 625 (1980).
- ¹⁵C. A. Truesdell and W. Noll, "The non-linear field theories of mechanics," in *Encyclopaedia of Physics* (Springer-Verlag, Berlin, 1965), Vol. III/3.
- ¹⁶L. G. Leal, *Laminar Flow and Convective Transport Processes* (Butterworth-Heinemann, Boston, 1992).
- ¹⁷R. G. Cox, "The deformation of a drop in a general time-dependent fluid flow," *J. Fluid Mech.* **37**, 601 (1969).
- ¹⁸G. Astarita and G. Marrucci, *Principles of Non-Newtonian Fluid Mechanics* (McGraw-Hill, Maidenhead, 1974).
- ¹⁹R. B. Bird, C. F. Curtiss, R. C. Armstrong, and O. Hassager, *Dynamics of Polymeric Liquids. Vol. 2: Kinetic Theory*, 2nd ed. (Wiley, New York, 1987).
- ²⁰S. Guido, F. Greco, and M. Villone, "Experimental determination of drop shape in slow steady shear flow," *J. Colloid Interface Sci.* **219**, 298 (1999).
- ²¹S. Guido and F. Greco, "Drop shape under slow steady shear flow and during relaxation. Experimental results and comparison with theory," *Rheol. Acta* **40**, 176 (2001).
- ²²F. Greco, "Single drop immersed in a moving fluid: Analytical solution for slow flows of second-order fluids," in *Proceedings of the XIIIth International Congress on Rheology II*, edited by D. M. Binding, N. E. Hudson, J. Mewis, J. M. Piau, C. J. S. Petrie, P. Townsend, M. H. Wagner, and K. Walters (Cambridge, United Kingdom, 2000), p. 348.