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Recriprocal theorem for concentric compound drops in arbitrary Stokes flows

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The Lorentz reciprocal theorem is generalized and applied to the study of the quasi-steady motion of a concentric spherical (CS) compound drop at zero Reynolds number. Using this result, the migration velocities of a force-free CS compound drop placed in a general ambient Stokes flow, as well as the forces on each drop when subjected to specified migration velocities, are calculated. The latter constitutes a generalization of Faxén's law to the case of a CS compound drop. Also some earlier results on the thermocapillary migration of such drops (Borhan *et al.* 1992) are rederived more simply and in greater generality.

1. Introduction

In this paper, we develop the Lorentz reciprocal theorem for a compound drop consisting of a spherical liquid drop placed in an unbounded fluid phase and containing a smaller droplet or gas bubble. We restrict our investigation to the low-Reynolds-number regime, with surface tension forces dominating the viscous forces as far as the deformations are concerned. The inner droplet is concentric with the outer drop, and all phases are immiscible with one another.

There has been renewed interest in the fluid dynamics of compound drops, after a relatively extended period of inactivity which followed the original experimental studies of Chambers & Kopac (1937) and Kopac & Chambers (1937). The recent interest is mostly due to the development of new applications of compound drops in a variety of processes such as artificial blood oxygenation (Li & Asher 1973), hydrocarbon separation (Li 1971), and prolongation of drug release (Brodin, Kavaliunas & Frank 1978). Whereas experimental studies on compound drops date back to Chambers & Kopac (1937), theoretical investigations of their motion are quite recent, as summarized in the review article by Johnson & Sadhal (1985). Most of the earlier analyses are concerned with the motion of these drops under gravity, with little or no inertial effects (Brunn & Roden 1985; Sadhal & Oguz 1985). Rushton & Davies (1983) examined the settling of encapsulated drops; Stone & Leal (1990) examined the behaviour of a compound drop in a general linear flow, with particular emphasis on the mechanisms leading to their breakup. More recently, prompted by the growing prospects for material processing in space, the thermocapillary motion of these drops has been studied (Shankar & Subramanian 1983; Morton, Subramanian &

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Balasubramanian 1990; Borhan, Haj-Hariri & Nadim 1992). The motion in these problems, wherein gravitational effects are absent, is driven by the imposition of a constant temperature gradient on the bulk fluid, which induces interfacial tension gradients on the drop and the droplet interfaces. These variations in interfacial tension result in surface velocities which, in turn, give rise to velocities in the bulk phases through viscous forces, leading to an eventual migration of the compound drop.

As was shown by Sadhal & Oguz (1985) for the motion of a compound drop under the action of gravity, the relative motion of the inner droplet with respect to the drop is very small for a wide range of densities and viscosities. This provides some justification for examining concentric geometries of the compound drop here, as well as in the previous studies by Shankar & Subramanian (1983), Morton *et al.* (1990), and Borhan *et al.* (1992). The primary result of most analytical investigations of the behaviour of compound drops has been the prediction of integral quantities such as the migration velocities or drag forces. However, details of the flow field have invariably had to be determined in order to calculate the desired integral quantities.

For problems with negligible advection of momentum and energy, the governing equations are linear and exactly solvable for the spherical geometries addressed in this work. However, through a generalization of the Lorentz reciprocal theorem, one can develop expressions for the desired integral quantities without the need to solve for the details of the flow. Details of derivation are presented in §2. Using these results, the migration velocities of a force-free concentric spherical (CS) compound drop, as well as the forces on the drop and the droplet moving with prescribed velocities in an arbitrary ambient Stokes flow, are calculated in §3. The latter result constitutes Faxén's law for a CS compound drop. Furthermore, as an example of the simplicity and generality of the recriprocal arguments presented here, some results on the thermocapillary migration of CS compound drops in the presence of surfactants (originally derived in Borhan *et al.* 1992) are rederived in §3. The derivations are much simplified and of greater generality. A brief discussion of the results follows in §4.

2. Recriprocal theorem

For low-Reynolds-number flows, there exists a reciprocal theorem due to Lorentz (cf. Happel & Brenner 1983) which often permits the calculation of gross quantities such as the net force on a drop or its force-free migration velocity, without actually solving for the flow field. In this section, we derive the relevant form of this reciprocal theorem for the case of a CS compound drop. Recent applications of this theorem to single-drop cases can be found in Rallison (1978), Leal (1980), and Haj-Hariri, Nadim & Borhan (1990).

Reciprocity principles are important tools in a number of other fields such as acoustics, electromagnetism, dynamics, and statics. The underlying requirement for the applicability of such principles is the existence of a self-adjoint mobility or admittance linear operator (matrix or differential). This condition is satisfied for the simple wave equation in acoustics (Pierce 1981) and electromagnetism, and for coupled oscillators with or without Rayleigh dissipation (Rayleigh 1873) including the static case of zero frequency. In acoustical applications one may interchange the locations of the source and receiver and still measure the same field, or one related to it, if the polarity of the source is altered along with its location. The applications in dynamics, statics, and electromagnetism are of the same nature.

For the case of Stokes flow, the reciprocity principle is obtained by considering two distinct sets of forces and velocities corresponding to the solution of two geometrically

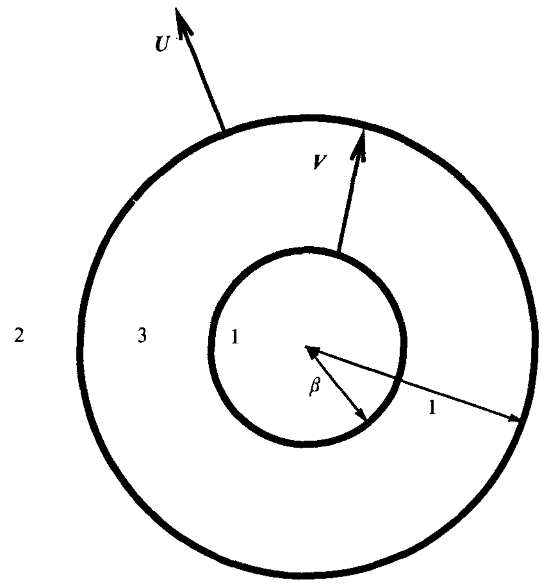

FIGURE 1. A concentric spherical (CS) compound drop.

identical problems (same geometry and governing equations, different boundary or forcing conditions). An alternative interpretation of this principle is one of conservation of a 'virtual energy' which for steady problems reduces to one of the balance of a 'virtual power flow'. The term 'virtual' is used to denote the formal nature of the entity: it is composed of forces from one experiment (solution) and velocities from another. In the case where both sets of data (solutions) correspond to the same experiment, this balance holds trivially. This notion of virtual power and the balance of its flux through the boundaries provides an alternative interpretation of the mathematical derivations which follow.

The problem of a CS compound drop translating in a quiescent fluid is now treated. Consider the following Stokes flow problem defined in the spherical geometry depicted in figure 1, with $\mathscr{D}_i$ and $\mathscr{S}_{ij}$ denoting the volume occupied by phase i and the interface between phases i and j, respectively:

$$\nabla^2 \boldsymbol{v}_i = \nabla p_i, \quad \nabla \cdot \boldsymbol{v}_i = 0 \quad \text{for} \quad \boldsymbol{r} \in \mathscr{D}_i, \tag{2.1 a, b}$$

subject to the boundary conditions

$$(\boldsymbol{v}_2, p_2) \rightarrow (-\boldsymbol{U}, \text{const.}), \quad \boldsymbol{r} \in \mathscr{S}_\infty, \tag{2.2}$$

and

$$\left.\begin{aligned} \boldsymbol{v}_i - \boldsymbol{v}_3 &= \boldsymbol{A}_{i3}(\boldsymbol{e}), & (2.3a)\\ \boldsymbol{e}\cdot\boldsymbol{v}_3 &= B_{i3}(\boldsymbol{e}) + \delta_{i1}\,\boldsymbol{e}\cdot(\boldsymbol{V}-\boldsymbol{U}), & (2.3b)\\ \boldsymbol{e}\cdot(\boldsymbol{\Pi}_3 - \lambda_i \boldsymbol{\Pi}_i)\cdot(\boldsymbol{I}-\boldsymbol{ee}) &= \boldsymbol{C}_{i3}(\boldsymbol{e}) & (2.3c) \end{aligned}\right\} \quad \boldsymbol{r} \in \mathscr{S}_{i3},$$

($i = 1, 2$). The surface $\mathscr{S}_\infty$ consists of a sphere at infinity, concentric with the surfaces $\mathscr{S}_{13}$ and $\mathscr{S}_{23}$. In these expressions, $\boldsymbol{v}_i$ and p_i denote the velocity and pressure fields in the three regions of a compound drop of outer radius unity and inner radius $\beta < 1$. By taking the above fields to be the *disturbance* quantities, the fluid far from the drop appears to be quiescent. However, the origin of the reference frame is fixed to the drop centre and translates with its unknown velocity $\boldsymbol{U}$ (cf. (2.2)). The inner droplet translates with the unknown velocity $\boldsymbol{V}$ which can be different from $\boldsymbol{U}$. The 'forcing' functions A_{i3}, B_{i3}, and C_{i3}, appearing in the boundary conditions at the interfaces, $\mathscr{S}_{i3}$, are arbitrary functions of position on those surfaces, as represented by their dependence on e, the unit vector in the radial direction – itself a function of the angular variables on the surface of a unit sphere. These forcing terms arise, for instance, when the problem is formulated for the disturbance fields, or when one seeks the leading correction to the flow for small perturbations away from the spherical geometry. The stress tensors $\boldsymbol{\Pi}_i$ have the usual Newtonian definitions (e.g. $\boldsymbol{\Pi}_i = -p_i \boldsymbol{I} + (\nabla \boldsymbol{v}_i + \nabla \boldsymbol{v}_i^{\dagger})$], and λ_i denotes the ratio of the viscosity of phase i to that of the intermediate phase, referred to as phase 3. The Newtonian nature of $\boldsymbol{\Pi}_i$ brings about the self-adjointness needed for the existence of a reciprocal principle. The notation and the non-dimensionalizations used in this paper are standard and conform to those of Haj-Hariri *et al.* (1990) and Nadim, Haj-Hariri & Borhan (1990). The pressure and stress in each phase are non-dimensionalized using the viscosity of that phase, so as to render (2.1) into the simplest form. For this derivation it is temporarily assumed that the drop and the droplet are force-free, namely

$$\int_{\mathscr{S}_{13}} \boldsymbol{e}\cdot\boldsymbol{\Pi}_3 \,\mathrm{d}S = \boldsymbol{0}, \quad \int_{\mathscr{S}_{23}} \boldsymbol{e}\cdot\boldsymbol{\Pi}_2 \,\mathrm{d}S = \boldsymbol{0}. \tag{2.4}$$

However, this assumption will be relaxed in obtaining Faxén's law. Boundary conditions (2.3) result from the continuity of velocity, the kinematic condition, and the tangential stress balance at the interfaces. The normal stress balance cannot be imposed because of the prespecification of the drop shapes; this condition can be used later to determine the corrections to the shapes by perturbation (e.g. Haj-Hariri *et al.* 1990). Given the forcing functions A_{i3}, B_{i3} and C_{i3}, the drop and the droplet achieve migration velocities $\boldsymbol{U}$ and $\boldsymbol{V}$, respectively, so as to satisfy (2.4).

As mentioned in Nadim *et al.* (1990), to derive the reciprocal theorem for the case of a single drop, one needs the flow field of a test problem, namely, that for uniform flow past a stationary drop. This known flow field, in conjunction with the boundary conditions for the problem of interest, leads to a simple expression for the migration velocity. In the present case there are two migration velocities that must be evaluated. Consequently, we will need the flow fields for two test problems; specifically, one corresponding to a CS compound drop in a quiescent fluid with the outer drop fixed and the inner droplet moving at $\boldsymbol{U}^*$, and the other to a uniform flow, $\boldsymbol{U}^*$, past a completely stationary CS compound drop. These test problems constitute two linearly independent solutions of the Stokes equations. Alternatively, the two problems are needed in order to uniquely determine the distribution of the power sinks on the two interfaces. Let us denote the velocity, pressure, and stress fields for the test problems by the symbols $\boldsymbol{u}$, q and $\boldsymbol{Q}$. Superscript $l (l = 1, 2)$ is used to distinguish the two test problems. The governing equations for these two problems are written with the aid of the Kronecker-delta, with the λ_i the same as those for the main problem:

$$\nabla^2 \boldsymbol{u}_i^l = \nabla q_i^l, \quad \nabla\cdot\boldsymbol{u}_i^l = 0 \quad \text{for} \quad \boldsymbol{r}\in\mathscr{D}_i, \tag{2.5a, b}$$

subject to the boundary conditions

$$(\boldsymbol{u}_2^l, q_2^l) \rightarrow (-\delta_{l2}\,\boldsymbol{U}^*, \text{const.}), \quad \boldsymbol{r}\in\mathscr{S}_\infty, \tag{2.6}$$

and

$$\left.\begin{aligned} &\boldsymbol{u}_i^l - \boldsymbol{u}_3^l = \boldsymbol{0}, && (2.7a)\\ &\boldsymbol{e}\cdot\boldsymbol{u}_3^l = \delta_{i1}\,\delta_{l1}\,\boldsymbol{e}\cdot\boldsymbol{U}^*, && (2.7b)\\ &\boldsymbol{e}\cdot(\boldsymbol{Q}_3^l - \lambda_i\,\boldsymbol{Q}_i^l)\cdot(\boldsymbol{I}-\boldsymbol{ee}) = \boldsymbol{0}, && (2.7c)\end{aligned}\right\} \quad \boldsymbol{r}\in\mathscr{S}_{i3},$$

where, for economy of notation, the constant velocity vector is denoted by the same symbol U^* in the two cases. Therefore, $l = 1$ corresponds to a stationary drop and a moving interior droplet with quiescent fluid at infinity, and $l = 2$ to a stationary CS compound drop with uniform flow at infinity, a problem solved by Rushton & Davies (1983). However, as will be shown in §3, by combining the two test problems in a reciprocal principle, the method becomes quite general and can be used to address problems that are considerably more involved than either of the test problems.

The solutions to these problems are written using Lamb's general solution in spherical geometries, as outlined in Appendix A. The general form of the solution is

$$\boldsymbol{u}_1^l = \hat{\boldsymbol{u}}^{\uparrow}(c_1^{l1}, c_2^{l1}) - \delta_{l2}\, \boldsymbol{U}^*, \quad q_1^l = \hat{q}^{\uparrow}(c_2^{l1}), \tag{2.8a}$$

$$\boldsymbol{u}_2^l = \hat{\boldsymbol{u}}^{\downarrow}(d_1^{l2}, d_2^{l2}) - \delta_{l2}\, \boldsymbol{U}^*, \quad q_2^l = \hat{q}^{\downarrow}(d_2^{l2}), \tag{2.8b}$$

$$\boldsymbol{u}_3^l = \hat{\boldsymbol{u}}^{\uparrow}(c_1^{l3}, c_2^{l3}) + \hat{\boldsymbol{u}}^{\downarrow}(d_1^{l3}, d_2^{l3}) - \delta_{l2}\, \boldsymbol{U}^*, \quad q_3^l = \hat{q}^{\uparrow}(c_2^{l3}) + \hat{q}^{\downarrow}(d_2^{l3}), \tag{2.8c}$$

where the superscripts $\uparrow$ and $\downarrow$, respectively denote algebraically growing and decaying behaviour in the radial coordinate, r. The second superscript on the constants c and d in (2.8) designates the fluid phase in the CS compound drop. The functional dependencies of $\hat{\boldsymbol{u}}$ and $\hat{q}$ on r and $\boldsymbol{e}(\boldsymbol{r} = r\boldsymbol{e})$ are implied but not shown explicitly. Their exact forms as well as the values of the constants c and d are presented in Appendix A. In the interest of brevity, the superscript l is suppressed hereafter unless it is needed explicitly. Using the expressions for $\boldsymbol{u}_i$, $\lambda_i \boldsymbol{e}\cdot\boldsymbol{Q}_i$, and $\boldsymbol{e}\cdot[\boldsymbol{Q}_3 - \lambda_i \mathbf{Q}_i]\cdot\boldsymbol{e}$ on the surfaces $\mathscr{S}_{i3}$, given at the end of Appendix A, it follows that the force on the drop is given as usual in terms of the stokeslet strength, $d_2^{(2)}$, as

$$\boldsymbol{P}_{23} = \int_{\mathscr{S}_{23}} \boldsymbol{e}\cdot\boldsymbol{Q}_2\, \mathrm{d}S = 8\pi \mathrm{d}_2^{(2)} \boldsymbol{U}^*. \tag{2.9}$$

In order to derive the reciprocal theorem for the CS compound drop, one starts with the low-Reynolds-number identities

$$\nabla\cdot(\boldsymbol{\Pi}_i\cdot\boldsymbol{u}_i^l) = \nabla\cdot(\boldsymbol{Q}_i^l\cdot\boldsymbol{v}_i), \tag{2.10}$$

which hold since $\boldsymbol{\Pi}$ and $\boldsymbol{Q}$, as well as the velocity fields, are solenoidal. Integrating (2.10) over $\mathscr{D}_i (i = 1, 2, 3)$, with $\mathscr{D}_2$ denoting the volume bounded by $\mathscr{S}_{23}$ and $\mathscr{S}_\infty$, and applying the Gauss divergence theorem results in

$$\int_{\mathscr{S}_\infty} \boldsymbol{e}\cdot\boldsymbol{\Pi}_2\cdot\boldsymbol{u}_2\, \mathrm{d}S - \int_{\mathscr{S}_{23}} \boldsymbol{e}\cdot\boldsymbol{\Pi}_2\cdot\boldsymbol{u}_2\, \mathrm{d}S = \int_{\mathscr{S}_\infty} \boldsymbol{e}\cdot\mathbf{Q}_2\cdot\boldsymbol{v}_2\, \mathrm{d}S - \int_{\mathscr{S}_{23}} \boldsymbol{e}\cdot\mathbf{Q}_2\cdot\boldsymbol{v}_2\, \mathrm{d}S, \tag{2.11a}$$

$$\int_{\mathscr{S}_{23}} \boldsymbol{e}\cdot\boldsymbol{\Pi}_3\cdot\boldsymbol{u}_3\, \mathrm{d}S - \int_{\mathscr{S}_{13}} \boldsymbol{e}\cdot\boldsymbol{\Pi}_3\cdot\boldsymbol{u}_3\, \mathrm{d}S = \int_{\mathscr{S}_{23}} \boldsymbol{e}\cdot\mathbf{Q}_3\cdot\boldsymbol{v}_3\, \mathrm{d}S - \int_{\mathscr{S}_{13}} \boldsymbol{e}\cdot\mathbf{Q}_3\cdot\boldsymbol{v}_3\, \mathrm{d}S, \tag{2.11b}$$

$$\int_{\mathscr{S}_{13}} \boldsymbol{e}\cdot\boldsymbol{\Pi}_1\cdot\boldsymbol{u}_1\, \mathrm{d}S = \int_{\mathscr{S}_{13}} \boldsymbol{e}\cdot\boldsymbol{Q}_1\cdot\boldsymbol{v}_1\, \mathrm{d}S. \tag{2.11c}$$

In the first term on the left-hand side of (2.11 *a*), $\boldsymbol{u}_2^l$ can be replaced by its constant value at infinity (cf. (2.6)), $\delta_{2l}\, \boldsymbol{U}^*$, and taken outside the integral. The remaining integral vanishes since the drop in the real problem is assumed to be force-free (cf. (2.1)–(2.3)). In §3 the case of non-vanishing force will be considered. Similarly, in the first term on the right-hand side of (2.11 *a*), $\boldsymbol{v}_2$ is replaced by the constant $-\boldsymbol{U}$ (cf. (2.2)), and since $\nabla\cdot\boldsymbol{Q}_2 = 0$ in $\mathscr{D}_3$, the remaining integral over $\mathscr{S}_\infty$ is recognized as the force on the CS compound drop of test problem l, $\boldsymbol{P}_{23}^l$, as given by (2.9). If both sides of (2.11 *a*) are multiplied by λ_2 and subtracted from (2.11 *b*), and those of (2.11 *c*) are multiplied by λ_1 and added to the result, it is found (after some simplification) that

$$\lambda_1 \boldsymbol{P}_{23}\cdot\boldsymbol{U}+\left[\int_{\mathscr{S}_{13}} \boldsymbol{e}\cdot[\boldsymbol{Q}_3-\lambda_1\boldsymbol{Q}_1]\cdot\boldsymbol{ee}\,\mathrm{d}S\right]\cdot(\boldsymbol{V}-\boldsymbol{U})$$

$$=\int_{\mathscr{S}_{23}}\{\boldsymbol{e}\cdot[\boldsymbol{Q}_3-\lambda_2\boldsymbol{Q}_2]\cdot\boldsymbol{e}B_{23}-\lambda_2\boldsymbol{e}\cdot\boldsymbol{Q}_2\cdot\boldsymbol{A}_{23}-\boldsymbol{C}_{23}\cdot\boldsymbol{u}_3\}\,\mathrm{d}S$$

$$-\int_{\mathscr{S}_{13}}\{\boldsymbol{e}\cdot[\boldsymbol{Q}_3-\lambda_1\boldsymbol{Q}_1]\cdot\boldsymbol{e}B_{13}-\lambda_1\boldsymbol{e}\cdot\boldsymbol{Q}_1\cdot\boldsymbol{A}_{13}-\boldsymbol{C}_{13}\cdot(\boldsymbol{u}_3-\delta_{l1}\boldsymbol{U}^*)\,\mathrm{d}S. \quad (2.12)$$

Using the interfacial conditions (2.3) and the force-free requirements (2.4), the integrals on the left-hand side of this equation can be simplified to yield the following system of equations for the two unknown migration velocities, $\boldsymbol{U}$ and $\boldsymbol{V}$:

$$\lambda_2 d_2^{(2)}\boldsymbol{U}+d_2^{(3)}(\boldsymbol{V}-\boldsymbol{U})=\frac{1}{8\pi}\left\{\int_{\mathscr{S}_{23}}\boldsymbol{K}_{23}\,\mathrm{d}S+\int_{\mathscr{S}_{13}}\boldsymbol{K}_{13}\,\mathrm{d}S\right\}, \quad (2.13)$$

where

$$\boldsymbol{K}_{13}=-2c_2^{(1)}\beta(\beta\boldsymbol{C}_{13}+9\lambda_1\boldsymbol{A}_{13})\cdot\boldsymbol{ee}+(c_1^{(1)}+4c_2^{(1)}\beta^2-1)$$
$$\times\boldsymbol{C}_{13}+6\lambda_1 c_2^{(1)}\beta\boldsymbol{A}_{13}-\frac{6d_2^{(3)}}{\beta^2}B_{13}\boldsymbol{e}, \quad (2.14a)$$

and

$$\boldsymbol{K}_{23}=(-\boldsymbol{C}_{23}+6\lambda_2\boldsymbol{A}_{23})\cdot[(3d_1^{(2)}+d_2^{(2)})\boldsymbol{ee}+d_1^{(2)}\boldsymbol{I}]$$
$$+(d_2^{(2)}+\delta_{2l})\boldsymbol{C}_{23}+6(d_2^{(3)}-\lambda_2 d_2^{(2)})B_{23}\boldsymbol{e}. \quad (2.14b)$$

The linearity of the equations in the arbitrary vector $\boldsymbol{U}^*$ has been exploited so as to eliminate this vector. The solution of the above system (for $l = 1,2$, which is a suppressed superscript on all coefficients) will yield the migration velocities for a wide range of problems in which the CS compound drop may deviate from its concentric configuration only at a very slow rate. This condition implies a vanishingly small relative migration velocity between the drop and the occluded droplet, consistent with the existing experimental results (e.g. Sadhal & Oguz 1985). The desired migration velocities are obtained without having to solve for the details of the flow field. Only two simple test problems needed to be solved in detail. All that is needed for studying new problems with the appropriate geometries is to determine the terms $\boldsymbol{A}_{i3}$, B_{i3}, and $\boldsymbol{C}_{i3}$ in the interfacial conditions (2.3) for the problem of interest. It can be shown that the constants d_2^{l2} and d_2^{l3}, appearing as the coefficients of our 2×2 algebraic system for $\boldsymbol{U}$ and $\boldsymbol{V}$, yield a non-vanishing Jacobian, thus ensuring uniqueness of the solution.

3. Results

3.1. *Force-free migration velocities*

The reciprocal argument is now used to determine the migration velocities of a CS compound drop placed in an arbitrary ambient Stokes field, $\boldsymbol{V}^\infty$, p^∞, satisfying the non-dimensional Stokes and continuity equations everywhere. The total velocity and pressure fields in the various phases, in the presence of the drop, are decomposed into the ambient field plus a disturbance, as follows:

$$\boldsymbol{v}_i=\boldsymbol{V}^\infty+\boldsymbol{v}_i', \quad p_i=p^\infty+p_i'. \quad (3.1a, b)$$

The disturbance quantities v_i' and p_i' satisfy the non-dimensionalized Stokes and continuity equations (cf. (2.1 *a*, *b*)), subject to the boundary conditions

$$(v_2', p_2') \to (-U, 0) \quad \text{as} \quad r \in \mathscr{S}_\infty, \tag{3.2}$$

and

$$\left.\begin{aligned} v_i' &= v_3', \\ e\cdot v_3' &= -e\cdot V^\infty + \delta_{i1}\, e\cdot(V-U), \end{aligned}\right\} \; r \in \mathscr{S}_{i3}. \qquad (3.3\,a,b)$$

The tangential-stress conditions at the interfaces are given as

$$e\cdot[\boldsymbol{\Pi}_3 - \lambda_i \boldsymbol{\Pi}_i]\cdot(\boldsymbol{I}-ee) = \boldsymbol{0} \quad \text{on} \quad r \in \mathscr{S}_{i3}. \tag{3.4}$$

Furthermore, force-free conditions are imposed on the drop and the droplet. Equation (3.4) can be written in terms of the disturbance quantities as

$$e\cdot[\boldsymbol{\Pi}_3' - \lambda_i \boldsymbol{\Pi}_i']\cdot(\boldsymbol{I}-ee) = (\lambda_i - 1)\, e\cdot\boldsymbol{\Pi}^\infty\cdot(\boldsymbol{I}-ee), \tag{3.5}$$

where $\boldsymbol{\Pi}^\infty$ is the dimensionless ambient stress tensor. These equations conform to the general form to which (2.13) applies, yielding the desired migration velocities. Noting the form of the right-hand side of (3.5), it is clear that the isotropic part of $\boldsymbol{\Pi}^\infty$ (i.e. p^∞) does not contribute to the final solution. Dropping the primes and performing some algebraic manipulations, one arrives at

$$\begin{aligned} 8\pi\lambda_2 d_2^{l2}\, U + 8\pi d_2^{l3}(V-U) = {} & (1-\lambda_2)(d_1^{l2} - d_2^{l2} - \delta_{l2}) \int_{\mathscr{S}_{23}} e\cdot[\nabla V^\infty + (\nabla V^\infty)^\dagger] \\ & \times (\boldsymbol{I}-ee)\,\mathrm{d}S + (\lambda_1 - 1)(c_1^{l1} + 4c_2^{l1}\beta^2 - 1) \\ & \times \int_{\mathscr{S}_{13}} e\cdot[\nabla V^\infty + (\nabla V^\infty)^\dagger]\cdot(\boldsymbol{I}-ee)\,\mathrm{d}S - 6(d_2^{l3} - \lambda_2 d_2^{l2}) \\ & \times \int_{\mathscr{S}_{23}} ee\cdot V^\infty\,\mathrm{d}S + \frac{6 d_2^{l3}}{\beta^2}\int_{\mathscr{S}_{13}} ee\cdot V^\infty\,\mathrm{d}S \end{aligned} \tag{3.6}$$

for $l = 1, 2$.

If the terms involving the ambient velocity are Taylor expanded about the origin, upon surface integration only a few terms remain nonzero, because V^∞ is a solution of the Stokes equations. Hence (3.6) is simplified to yield the migration velocity of the outer drop as

$$U = V_o^\infty - \frac{2(\lambda_1 - 1)\beta^5 + (3\lambda_1 + 2)}{2E_{uv}} \nabla^2 V_o^\infty, \tag{3.7}$$

$$E_{uv} \equiv 6\beta^5(\lambda_1 - 1)(\lambda_2 - 1) - (3\lambda_1 + 2)(2\lambda_2 + 3), \tag{3.8}$$

where subscript 'o' indicates that the corresponding term is evaluated at the drop origin. In the single-drop limit, $\beta \to 0$, (3.7) reduces to the well-known expression

$$\lim_{\beta\to 0} U = V_o^\infty + \frac{1}{2(2\lambda_2 + 3)} \nabla^2 V_o^\infty. \tag{3.9}$$

The migration velocity of the interior droplet is in turn given by

$$V = V_o^\infty - \frac{\lambda_1 \lambda_2 \beta^2}{2E_{uv}} \nabla^2 V_o^\infty. \tag{3.10}$$

Expressions (3.7) and (3.10) are compact representations for the migration velocities of a force-free CS compound drop placed in an arbitrary ambient Stokes flow field. These expressions are valid so long as $|U-V|/|U| \ll 1$ so that the concentric geometry is preserved.

3.2. *Faxén's law*

In this subsection, the forces on the drop and the droplet resulting from the prescribed motion of a CS compound drop with velocities U and V in a general Stokes flow are calculated. The force on each drop is given by

$$\boldsymbol{F}_{13} = \int_{\mathscr{S}_{13}} \boldsymbol{e}\cdot\boldsymbol{\Pi}_3\,\mathrm{d}S, \quad \boldsymbol{F}_{23} = \int_{\mathscr{S}_{23}} \boldsymbol{e}\cdot\boldsymbol{\Pi}_2\,\mathrm{d}S. \tag{3.11}$$

When a decomposition similar to that in §3.1 (cf. equation (3.1)) is again used, the part of the stress tensor due to the ambient flow will have no contribution to the force integrals of (3.11) since $\boldsymbol{\Pi}^{\infty}$ is solenoidal. Thus the forces can be simply calculated from the disturbance parts of the stress tensors.

The governing equations are the same as those in §3.1 above, with the exception that U and V are now prescribed rather than being unknown. A derivation very similar to that in §2 can be performed for the forces instead of the migration velocities. The derivation is modified only by its treatment of the integrals at infinity, where some terms that vanished before (in (2.11 *a*)) are now retained. The resulting expressions for the forces are

$$\boldsymbol{F}_{i3} = -\frac{8\pi}{\lambda_2\delta_{i2}+\delta_{i1}}\{(-\lambda_2 d_2^{i2}+d_2^{i3})\,\boldsymbol{U}-d_2^{i3}\boldsymbol{V}+\lambda_2 d_2^{i2}\,\boldsymbol{V}_{\mathrm{o}}^{\infty}+\tfrac{1}{10}[(\lambda_2-1)(2d_2^{i2}+\tfrac{3}{2}\delta_{i2}) \\ +(\lambda_2 d_2^{i2}-d_2^{i3})+\beta^2 d_2^{i3}+\beta^3(\lambda_1-1)(c_1^{i1}+4c_2^{i1}\beta^2-1)]\,\nabla^2\boldsymbol{V}_{\mathrm{o}}^{\infty}\} \tag{3.12}$$

for $i=1,2$. The above expressions for the forces on the drop and the droplet can formally be written as

$$\boldsymbol{F}_{i3} = F_U^{i3}\,\boldsymbol{U}+F_V^{i3}\,\boldsymbol{V}+F_{V\infty}^{i3}\,\boldsymbol{V}_{\mathrm{o}}^{\infty}+F_{\nabla^2 V^\infty}^{i3}\,\nabla^2\boldsymbol{V}_{\mathrm{o}}^{\infty}, \quad i=1,2, \tag{3.13}$$

with expressions for the various F^{i3} included in Appendix B. To check the validity of these expressions, we examine them in the limit as the radius of the interior droplet vanishes. It is readily observed that all contributions to the force on interface $\mathscr{S}_{13}$ vanish, and the components of the force on interface $\mathscr{S}_{23}$ become independent of the viscosity of the droplet, λ_1. It is also found that the component F_V^{23} vanishes altogether. If the viscosity of the drop is allowed to approach infinity, so that the drop represents a solid sphere, the expression for the force on this infinitely viscous drop becomes

$$\boldsymbol{F}_{\mathrm{sph}} = \lim_{\beta\to 0,\,\lambda_2\to 0} \boldsymbol{F}_{23} = 6\pi\{1+\tfrac{1}{6}\nabla^2\}\,\boldsymbol{V}_{\mathrm{o}}^{\infty}, \tag{3.14}$$

which is the well-known Faxén's law for a solid sphere in an arbitrary Stokes flow (cf. Kim & Karrila 1991). Moreover, the force-free migration velocities (cf. §3.1) are recovered by selecting U and V in (3.13) so as to nullify the forces on the compound drop.

3.3. *Thermocapillary migration*

In this subsection, the thermocapillary migration velocities of a CS compound drop, and corrections thereto in the presence of surface-active impurities are derived using the reciprocal theorem. The results are compared with those originally derived in Morton *et al.* (1990) and Borhan *et al.* (1992). The equations governing the thermocapillary migration of a CS compound drop in the presence of surfactants are presented below. Upon identification of the applicable $\boldsymbol{A}_{i3}$, B_{i3}, and $\boldsymbol{C}_{i3}$ (cf. (2.3)), expression (2.13) is used to determine the migration velocities $\boldsymbol{U}$ and $\boldsymbol{V}$.

The thermocapillary motion of the drop is due to the dependence of surface tension on temperature, and is modified by its additional dependence on surfactant concentration. The velocity and pressure fields, $\boldsymbol{v}_i$ and p_i, of a CS compound drop satisfy the non-dimensional Stokes and continuity equations (2.1) subject to the boundary conditions (2.2) and (2.3). For this case, the functions $\boldsymbol{A}_{i3}$, B_{i3}, and $\boldsymbol{C}_{i3}$ $(i = 1, 2)$, appearing in the interfacial conditions (2.3*a–c*), are given by

$$\boldsymbol{A}_{i3}(\boldsymbol{e}) = \boldsymbol{0}, \tag{3.15a}$$

$$B_{i3}(\boldsymbol{e}) = 0, \tag{3.15b}$$

$$\boldsymbol{C}_{i3}(\boldsymbol{e}) = (-1)^{(i+1)}[(\delta_{i1}+\gamma\delta_{i2})\boldsymbol{\nabla}_s T_{i3}+(\delta_{i1}+\alpha\delta_{i2})\epsilon\boldsymbol{\nabla}_s \Gamma_{i3}]\cdot(\boldsymbol{I}-\boldsymbol{ee}), \tag{3.15c}$$

where T_{i3} and Γ_{i3} represent the temperature and surfactant concentrations, respectively, on interfaces $\mathscr{S}_{i3}(i = 1, 2)$. The symbols α, γ, and ϵ are defined by

$$\alpha = \frac{1}{\sigma_o}\left(\frac{\partial\sigma}{\partial\Gamma}\right)_{\Gamma_o}, \quad \gamma = \frac{1}{\sigma_o}\left(\frac{\partial\sigma}{\partial T}\right)_{T_o}, \quad \epsilon = \frac{\alpha\Gamma_o}{\gamma T_o}. \tag{3.16}$$

In the above, σ is the surface tension and the subscript 'o' represents evaluation at the reference temperature and surfactant concentration, T_o and Γ_o, respectively.

The reciprocal theorem is used to determine the thermocapillary migration velocities of the droplet and the drop in the absence ($\epsilon = 0$) as well as in the presence of trace amounts of surfactant ($0 < \epsilon \ll 1$). For $\epsilon = 0$, the 'forcing' function $\boldsymbol{C}_{i3}$ takes the form

$$\boldsymbol{C}_{i3} = C_{i3}(\boldsymbol{I}-\boldsymbol{ee})\cdot\boldsymbol{G}, \tag{3.17}$$

where $\boldsymbol{G}$ is the constant temperature gradient, and the constants C_{i3} are related to the surface gradients of the temperature field on $\mathscr{S}_{i3}(i = 1, 2)$, given explicitly in Appendix B. Equation (2.13) immediately provides the migration velocities

$$\boldsymbol{U} = -\frac{10\beta^4 C_{13}+2[3(\lambda_1-1)\beta^5-(3\lambda_1+2)]C_{23}}{3E_{uv}}\hat{\boldsymbol{G}}, \tag{3.18a}$$

$$\boldsymbol{V} = -\frac{5[3\lambda_1(\beta^2-1)-2]C_{23}+[6(\lambda_2-1)\beta^6-10(\lambda_2-1)\beta^4+2(2\lambda_2+3)\beta]C_{13}}{2E_{uv}}\hat{\boldsymbol{G}}, \tag{3.18b}$$

with E_{uv} given in (3.8), and $\hat{\boldsymbol{G}} \equiv \boldsymbol{G}/|\boldsymbol{G}|$. These results are in complete agreement with those of Morton *et al.* (1990).

To determine the corrections to the thermocapillary migration velocities of the CS compound drop in the presence of bulk-insoluble surfactants, the surfactant concentrations Γ_{i3} from Borhan *et al.* (1992) are used. These concentration profiles were obtained by solving the surface convective–diffusion equation using the leading-order surface velocities which are known for $\epsilon = 0$. The surfactant distribution on the interface is given by

$$\Gamma_{i3} = \frac{\mathscr{P}_i \exp[(-1)^{i+1}\mathscr{P}_i \boldsymbol{e}\cdot\hat{\boldsymbol{G}}]}{\sinh\mathscr{P}_i}, \tag{3.19}$$

with the $\mathscr{P}_i(i = 1, 2)$ denoting the surface Péclet numbers for surfactant transport (cf. Borhan *et al.* 1992). The $O(\epsilon)$ correction fields satisfy equations similar to the 'clean' thermocapillary motion above, except that the functions C_{i3} now result from the surface gradient of the surfactant concentrations (3.19). Expressions for C_{i3} for this problem turn out to be

$$C_{23} = -\nabla_s \Gamma_{23} = \frac{\mathscr{P}_2^2}{\sinh^2 \mathscr{P}_2} \exp(-\mathscr{P}_2 \boldsymbol{e}\cdot\hat{\boldsymbol{G}})(\boldsymbol{I}-\boldsymbol{ee})\cdot\hat{\boldsymbol{G}}, \tag{3.20a}$$

$$C_{13} = \alpha\nabla_s \Gamma_{13} = \frac{\alpha\mathscr{P}_2^2}{\sinh^2 \mathscr{P}_1} \exp(\mathscr{P}_1 \boldsymbol{e}\cdot\hat{\boldsymbol{G}})(\boldsymbol{I}-\boldsymbol{ee})\cdot\hat{\boldsymbol{G}}. \tag{3.20b}$$

Carrying out the requisite integrations in (2.13), the $O(\epsilon)$ correction to the migration velocity of the drop, U, is found to be

$$U_\Gamma = -\frac{10\beta^4\alpha_1[1/\mathscr{P}_1 - \coth\mathscr{P}_1] + 2[3(\lambda_1-1)\beta^5-(3\lambda_1+2)][\coth\mathscr{P}_2 - 1/\mathscr{P}_2]}{3E_{uv}}\hat{\boldsymbol{G}}, \tag{3.21}$$

with a similar expression for V_Γ; one related to (3.18*b*) in the same way that (3.21) is related to (3.18*a*). The advantage of using the reciprocal theorem for calculating the correction to the migration velocity is that it provides an expression valid for all Péclet numbers, whereas the original perturbation solution of Borhan *et al.* (1992) yields the correction for small Péclet numbers only. The solution obtained here agrees with the perturbation results as $\mathscr{P}_i$ tends to zero.

4. Discussion

We have applied the Lorentz reciprocal theorem to the case of a concentric spherical compound drop in quasi-steady motion. As mentioned in the introduction, this is not a contrived example in the sense that the concentric geometry of the compound drops has been shown in the literature to be a prevalent one (e.g. Sadhal & Oguz 1985). Applications to forced and force-free motions of a CS compound drop in an arbitrary ambient Stokes flow were demonstrated. The forced-drop results constitute an extension of Faxén's laws to the case of CS compound drops. Applications to the thermocapillary migration of compound drops in the presence and absence of surface–active impurities are shown to generalize the results derived earlier by Borhan *et al.* (1992). This generalization was mainly in the form of removing the small surface Péclet number restriction. The main advantage of such reciprocal arguments is that only one or two test problems, which are usually straightforward, need be solved in detail. Once the solutions to the test problems have been obtained, a whole host of geometrically compatible problems can be studied and integral quantities such as migration velocities or forces can be calculated elegantly and with relative ease.

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Appendix A. The two test problems

As mentioned in the derivation of §2, two 'linearly independent' solutions of the Stokes equations are necessary for the CS compound drop geometry in order to determine the unknown quantities of interest (velocities or forces). The two problems selected in the current investigation correspond to (2.5)–(2.7) and can be solved using a vector form of Lamb's general solution (Hinch 1988). The general expressions for the velocity and pressure fields are

$$\boldsymbol{u} = \nabla\phi + \boldsymbol{r}\wedge\nabla\psi + \nabla(\boldsymbol{r}\cdot\boldsymbol{A}) - 2\boldsymbol{A}, \quad q = 2\nabla\cdot\boldsymbol{A}, \tag{A 1}$$

where the fields ϕ, ψ, and $\boldsymbol{A}$ are all harmonic, and $\boldsymbol{A}$ is non-solenoidal. Since the problem is linear, the above harmonic functions must all be linear in the forcing vector, $\boldsymbol{U}^*$. Clearly ψ will not contribute since it is a pseudoscalar and cannot be constructed from the true vector $\boldsymbol{U}^*$. The solution in the exterior fluid phase, labelled 2 in figure 1, is constructed using exterior spherical harmonics, whereas inside the inner droplet, labelled 1 in figure 1, the interior spherical harmonics are used. In the intermediate region, 3, a combination of both types of solution will be needed. The exterior and interior forms of solution each involve two scalar constants. Specifically, for the exterior solution

$$\phi = (d_1/r^2)\boldsymbol{e}\cdot\boldsymbol{U}^*, \quad \psi = 0, \quad \boldsymbol{A} = d_2(1/r)\,\boldsymbol{U}^*, \tag{A 2}$$

yielding

$$\boldsymbol{u} = \hat{\boldsymbol{u}}^{\downarrow}(d_1, d_2) \equiv \left\{-\left(\frac{3d_1}{r^3}+\frac{d_2}{r}\right)\boldsymbol{ee} + \left(\frac{d_1}{r^3}-\frac{d_2}{r}\right)\boldsymbol{I}\right\}\cdot\boldsymbol{U}^* \tag{A 3a}$$

and

$$q = \hat{q}^{\downarrow}(d_1, d_2) \equiv -(2d_2/r^2)\boldsymbol{e}\cdot\boldsymbol{U}^*, \tag{A 3b}$$

whereas for the interior

$$\phi = c_1 r\boldsymbol{e}\cdot\boldsymbol{U}^*, \quad \psi = 0, \quad \boldsymbol{A} = c_2 r^2[3\boldsymbol{ee}-\boldsymbol{I}]\cdot\boldsymbol{U}^*, \tag{A 4}$$

yielding

$$\boldsymbol{u} = \hat{\boldsymbol{u}}^{\uparrow}(c_1, c_2) \equiv \{-2c_2 r^2\boldsymbol{ee} + (c_1 + 4c_2 r^2)\boldsymbol{I}\}\cdot\boldsymbol{U}^* \tag{A 5a}$$

and

$$q = \hat{q}^{\uparrow}(c_1, c_2) \equiv 20c_2 r\boldsymbol{e}\cdot\boldsymbol{U}^*. \tag{A 5b}$$

The constants used in conjunction with the growing solutions are c_i^{ls} and those with the decaying solutions are d_i^{ls}, where $i(=1,2)$ is a counter for the constants, $l(=1,2)$ denotes the test problem being studied, and $s(=1,2,3)$ is the fluid phase indicator.

Satisfaction of the boundary conditions (2.6) and (2.7) yields the following 8×8 algebraic system for the constants c and d:

$$-2c_2^{(3)} + d_2^{(2)} + 3d_1^{(2)} - d_2^{(3)} - 3d_1^{(3)} = 0, \tag{A 6a}$$

$$4c_2^{(3)} + c_1^{(3)} + d_2^{(2)} - d_1^{(2)} - d_2^{(3)} + d_1^{(3)} = 0, \tag{A 6b}$$

$$-2\beta^2 c_2^{(3)} + 2\beta^2 c_2^{(1)} - d_2^{(3)}/\beta - 3d_1^{(3)}/\beta^3 = 0, \tag{A 6c}$$

$$4\beta^2 c_2^{(3)} + c_1^{(3)} - 4\beta^2 c_2^{(1)} - c_1^{(1)} - d_2^{(3)}/\beta + d_1^{(3)}/\beta^3 = 0, \tag{A 6d}$$

$$-2d_2^{(2)} - 2d_1^{(2)} = \delta_{l2}, \quad 2\beta^2 c_2^{(1)} + c_1^{(1)} = 1, \tag{A 6e,f}$$

$$\beta c_2^{(3)} - \lambda_1\beta c_2^{(1)} - \frac{d_1^{(3)}}{\beta^4} = 0, \quad c_2^{(3)} + \lambda_2 d_1^{(2)} - d_1^{(3)} = 0. \tag{A 6g, h}$$

The solution to this algebraic system of equations can be readily obtained using common symbolic algebra software. Some of the constants have rather cumbersome representations; however, it is remarkable that their combinations resulting in physical quantities of interest, such as forces or migration velocities, are relatively compact.

The following expressions are needed for the derivations presented in §2. On $\mathscr{S}_{13}$:

$$\boldsymbol{u}_1 = \boldsymbol{u}_3 \equiv \boldsymbol{u}_{13} = \{2c_2^{(1)}\beta^2\boldsymbol{ee} + (c_1^{(1)} + 4c_2^{(1)}\beta^2 - \delta_{2l})\boldsymbol{I}\}\cdot\boldsymbol{U}^*, \tag{A 7a}$$

$$\lambda_1\boldsymbol{e}\cdot\boldsymbol{Q}_1 = \lambda_1\{-q_{01}\boldsymbol{e} + 6(-3c_2^{(1)}\beta\boldsymbol{ee} + c_2^{(1)}\beta\boldsymbol{I})\cdot\boldsymbol{U}^*\}, \tag{A 7b}$$

$$\boldsymbol{e}\cdot[\boldsymbol{Q}_3 - \lambda_1\boldsymbol{Q}_1]\cdot\boldsymbol{e} = -(q_{03} - \lambda_1 q_{01}) + 6(d_2^{(3)}/\beta^2)\boldsymbol{e}\cdot\boldsymbol{U}^*; \tag{A 7c}$$

and on $\mathscr{S}_{23}$:

$$u_2 = u_3 \equiv u_{23} = \{-(3d_1^{(2)}+d_2^{(2)})\,ee+(d_1^{(2)}-d_2^{(2)}-\delta_{2l})\,I\}\cdot U^*, \qquad (\mathrm{A}\,8a)$$

$$\lambda_2\,e\cdot Q_2 = \lambda_2\{-q_{o2}\,e+6[(3d_1^{(2)}+d_2^{(2)})\,ee-d_1^{(2)}\,I]\cdot U^*\}, \qquad (\mathrm{A}\,8b)$$

$$e\cdot[Q_3-\lambda_2\,Q_2]\cdot e = -(q_{o3}-\lambda_2\,q_{o2})+6(d_2^{(3)}-\lambda_2\,d_2^{(2)})\,e\cdot U^*. \qquad (\mathrm{A}\,8c)$$

In (A 7) and (A 8), $q_{oi}(i=1,2,3)$ denote the constant part of the pressure in phase i for the test problem l. The above relations turn out to be the only results required in the application of the reciprocal theorem to the CS compound drop.

Appendix B. Definition of various constants

The forces on the drop and the droplet are given in (3.13) as

$$F_{i3} = F_U^{i3}\,U+F_V^{i3}\,V+F_{V^\infty}^{i3}\,V_o^\infty+F_{\nabla^2V^\infty}^{i3}\,\nabla^2 V_o^\infty, \quad i=1,2, \qquad (\mathrm{B}\,1)$$

where for the drop–droplet interface, $\mathscr{S}_{13}$:

$$F_U^{13} = -[4(\lambda_1-1)(\lambda_2-1)\beta^6+5\lambda_1\lambda_2\beta^3-(3\lambda_1+2)(3\lambda_2+2)\beta]/2E_f^{(1)}, \qquad (\mathrm{B}\,2a)$$

$$F_V^{13} = [(3\lambda_2-2)(\lambda_1-1)\beta^6-(3\lambda_1+2)(\lambda_2+1)\beta]/E_f^{(1)}, \qquad (\mathrm{B}\,2b)$$

$$F_{V^\infty}^{13} = -[2\lambda_2(\lambda_1-1)(\beta^5+\beta^4+\beta^3)-\lambda_2(3\lambda_1+2)(\beta^2+\beta)]/E_f^{(2)}, \qquad (\mathrm{B}\,2c)$$

$$F_{\nabla^2V^\infty}^{13} = -\tfrac{1}{2}F_{V^\infty}^{13}, \qquad (\mathrm{B}\,2d)$$

with

$$E_f^{(1)} = 4(\lambda_1-1)(\lambda_2-1)\beta^6+(3\lambda_1-2)(2-3\lambda_2)\beta^5+10\lambda_1\lambda_2\beta^3 -(3\lambda_1+2)(3\lambda_2+2)\beta+4(\lambda_1+1)(\lambda_2+1), \qquad (\mathrm{B}\,2e)$$

$$E_f^{(2)} = 8(\lambda_1-1)(\lambda_2-1)\beta^5+2[(2-5\lambda_1)\lambda_2+2\lambda_1](\beta^4+\beta^3) +2[(5\lambda_1+2)\lambda_2+2\lambda_1](\beta^2+\beta)+8(\lambda_1+1)(\lambda_2+1), \qquad (\mathrm{B}\,2f)$$

while on the drop–exterior interface, $\mathscr{S}_{23}$:

$$F_U^{23} = -\{4(\lambda_1-1)(\lambda_2-1)\beta^5+(2-5\lambda_1)(\lambda_2-1)(\beta^4+\beta^3) +[(5\lambda_1+2)\lambda_2-2](\beta^2+\beta)-2(\lambda_1+1)(2\lambda_2+3)\}/E_f^{(2)}, \qquad (\mathrm{B}\,3a)$$

$$F_V^{23} = [2(\lambda_1-1)(\beta^5+\beta^4+\beta^3)-(3\lambda_1+1)(\beta^2+\beta)]/E_f^{(2)}, \qquad (\mathrm{B}\,3b)$$

$$F_{V^\infty}^{23} = \{2(\lambda_1-1)(2\lambda_2-3)\beta^4-(\lambda_1+2)(\lambda_2-3)\beta^3+6(1-\lambda_1\lambda_2)\beta^2 +[(2-\lambda_1)\lambda_2+3\lambda_1+6]\beta+2(\lambda_1+1)(2\lambda_2+3)\}/E_F^{(3)}, \qquad (\mathrm{B}\,3c)$$

$$F_{\nabla^2V^\infty}^{23} = -[2(\lambda_1-1)\beta^4+(\lambda_1-2)\beta^3-2\beta^2-(\lambda_1+2)\beta-2(\lambda_1+1)]/2E_f^{(3)}, \qquad (\mathrm{B}\,3d)$$

with

$$E_f^{(3)} = 8(\lambda_1-1)(\lambda_2-1)\beta^4-2(\lambda_1+2)(\lambda_2-2)\beta^3+4(2-3\lambda_1\lambda_2)\beta^2 +2(2-\lambda_1)(\lambda_2+2)\beta+8(\lambda_1+1)(\lambda_2+2). \qquad (\mathrm{B}\,3e)$$

The expressions below define the C_{i3} appearing in §3.3:

$$C_{13} = -\frac{9\kappa_{23}}{E_T}, \quad C_{23} = -\left(1+\frac{\beta^3(1-\kappa_{13})(2+\kappa_{23})+(2+\kappa_{13})(\kappa_{23}-1)}{E_T}\right), \qquad (B\,4a,b)$$

with

$$E_T = 2\beta^3(1-\kappa_{13})(1-\kappa_{23})-(2+\kappa_{13})(2\kappa_{23}+1), \qquad (\mathrm{B}\,5)$$

where κ_{i3} is the ratio of the thermal conductivity of phase i to that of phase 3.

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