

# Thermocapillary migration of a bidisperse suspension of bubbles

By Y. WANG<sup>1</sup>, R. MAURI<sup>2</sup> AND A. ACRIVOS<sup>1</sup>

<sup>1</sup> Levich Institute, T-1M, City College of CUNY, New York, NY 10031, USA

<sup>2</sup> Department of Chemical Engineering, City College of CUNY, New York, NY 10031, USA

(Received 24 May 1993 and in revised form 18 August 1993)

We consider the thermocapillary motion of a well-mixed suspension of non-conducting spherical bubbles of negligible viscosity in a viscous conducting liquid under conditions of vanishingly small Reynolds and Marangoni numbers. Recently, Acrivos, Jeffrey & Saville (1990) showed that when all the bubbles are of identical size, the ensemble-averaged migration velocity  $\bar{U}_1$  of a test bubble of radius  $a_1$  within the suspension equals  $U_1^{(0)}[1 - \frac{3}{2}c_1 + O(c_1^2)]$ , where  $c_1$  is the volume fraction of the bubbles and  $U_1^{(0)}$  is the thermocapillary velocity of a single bubble given by Young, Goldstein & Block (1959). Here we extend this result to a bi-disperse suspension containing bubbles of radii  $a_1$  and  $a_2 \equiv \lambda a_1$  in which case  $\bar{U}_1 = U_1^{(0)}[1 - \frac{3}{2}c_1 - S(\lambda)c_2 + \dots]$ , where  $c_1$  and  $c_2$  are the corresponding volume fractions of the two sets of bubbles. Values for  $S(\lambda)$  are presented for some typical size ratios  $\lambda$ , and asymptotic expressions for  $S(\lambda)$  are derived for  $\lambda \rightarrow 0$  and for  $\lambda \rightarrow \infty$ .

---

## 1. Introduction

A cloud of bubbles suspended in a viscous liquid of non-uniform temperature will move towards the hotter fluid, owing to the dependence of surface tension on temperature. In addition to its importance from a fundamental point of view, this effect has taken on a new practical significance in that, under near-weightless conditions, it offers the only technique presently available for removing unwanted gas bubbles from a liquid solution. This in turn is a crucial step in the manufacturing process of ultra-high-purity materials in outer space.

The thermocapillary motion of a single drop having arbitrary values of its thermal conductivity and viscosity relative to those of the ambient liquid was studied by Young, Goldstein & Block (1959) under conditions of vanishingly small Reynolds and Marangoni numbers. More recently, that study was extended to the case of two bubbles or drops by, among others, Meyyappan & Subramanian (1984) and Anderson (1985) via the direct reflection method, and by Satrape (1992) using the method of twin multipole expansions, all of whom showed that, when the surface tension is high enough to keep them spherical, two equi-sized bubbles move with the same velocity as one isolated bubble. Acrivos, Jeffrey & Saville (1990) extended this result by proving that it continues to hold for any number of equi-sized bubbles. Moreover, those authors showed that the velocity field is irrotational and that its velocity potential is uniquely related to the temperature, so that the hydrodynamic and thermal two-body interactions exactly cancel each other.

On the other hand, when the two bubbles have different radii, the hydrodynamic and thermal effects no longer cancel each other so that the particle velocities (now different

from each other) will depend on the distance between the two bubbles, their orientation relative to the direction of the applied temperature gradient and on the ratio of their radii.

In this paper we wish to calculate the average velocity of a test bubble in a dilute suspension of bubbles having a different size. The renormalization technique developed by Jeffrey (1973) is applied to this problem, extending the result of Acrivos *et al.* (1990), who considered monodisperse suspensions. The paper is organized as follows. After formulating the problem in §2, the mobility functions for two bubbles with arbitrary size ratio and arbitrary orientation relative to the imposed temperature gradient are obtained in §3 using the twin multipole expansion method developed by Jeffrey & Onishi (1984). These mobility functions, expanded in terms of the interparticle distance and size ratio, are then employed in §4 to find the average bubble velocity by making use of a renormalization technique described by Acrivos *et al.* (1990). Finally in §5, asymptotic expressions for the average bubble velocity are obtained in the limiting cases where the size ratio is either very large or very small.

## 2. The governing equations and a relation for a special case

A cloud of  $N$  bubbles suspended in an unbounded fluid of viscosity  $\mu$ , density  $\rho$  and thermal diffusivity  $\bar{\alpha}$  will move under the influence of a non-uniform ambient temperature field  $T_\infty$ , due to the temperature dependence of the surface tension of the bubble–fluid interface. We assume that  $\gamma_i$ , the surface tension of any bubble  $i$  ( $i = 1, 2, \dots, N$ ), decreases linearly with the temperature  $T$  and is large enough to keep each bubble spherical with radius  $a_i$ . We also suppose that both the Reynolds number  $Re$  and the Marangoni number  $Ma$  are small, with  $Re = \rho a_i U / \mu$  and  $Ma = a_i U / \bar{\alpha}$ , where  $U$  denotes a characteristic velocity.

For the quasi-steady-state case being considered here, the governing equation and boundary conditions for the temperature field  $T$  are given by

$$\nabla^2 T = 0, \quad (1)$$

$$T \rightarrow T_\infty \quad \text{as} \quad \rho_i \rightarrow \infty, \quad (2)$$

$$\mathbf{n}_i \cdot \nabla T = 0 \quad \text{on} \quad \rho_i = a_i, \quad (3)$$

where  $\rho_i = |\boldsymbol{\rho}_i|$ , with  $\boldsymbol{\rho}_i$  denoting the position vector referred to the centre of the  $i$ th bubble,  $T_\infty$  is the unperturbed temperature field which also satisfies Laplace's equation, while  $\mathbf{n}_i$  is a unit vector normal to the surface of the  $i$ th bubble.

Similarly, the velocity field  $\mathbf{u}$  and pressure field  $p$  satisfy

$$\nabla^2 \mathbf{u} = \frac{1}{\mu} \nabla p, \quad (4)$$

$$\nabla \cdot \mathbf{u} = 0, \quad (5)$$

$$\mathbf{u} \rightarrow \mathbf{u}_\infty \quad \text{as} \quad \rho_i \rightarrow \infty, \quad (6)$$

$$\mathbf{n}_i \cdot \mathbf{u} = \mathbf{n}_i \cdot \mathbf{U}_i \quad \text{on} \quad \rho_i = a_i, \quad (7)$$

$$\mathbf{n}_i \cdot \boldsymbol{\sigma} \cdot (\mathbf{I} - \mathbf{n}_i \mathbf{n}_i) = \left( -\frac{d\gamma_i}{dT} \right) (\mathbf{I} - \mathbf{n}_i \mathbf{n}_i) \cdot \nabla T \quad \text{on} \quad \rho_i = a_i, \quad (8)$$

where  $\boldsymbol{\sigma}$  is the stress tensor,  $\mathbf{u}_\infty$  is the unperturbed velocity field which also satisfies (4) and (5), while  $\mathbf{U}_i$  is the velocity of the  $i$ th bubble. We further require that the bubbles be force-free, i.e.

$$\mathbf{F}_i = 0.$$

Note that the corresponding torque-free condition is automatically satisfied in view of the expression, given by (8), for the jump in shear stress along the surface of each bubble.

From (1)–(8) it is evident that, in the absence of convection, the transport of energy is independent of that of momentum. In fact, the temperature distribution can first be determined through (1)–(3), and the boundary value problem (4)–(8) can subsequently be solved to find the flow field.

The case of a single bubble suspended in an unbounded fluid in the presence of a constant ambient temperature gradient, i.e.  $\nabla T_\infty = \mathbf{H}$ , was first solved by Young *et al.* (1959), who found that the thermocapillary velocity of an isolated bubble is given by

$$\mathbf{U}_i^{(0)} = \frac{a_i}{2\mu} \left( -\frac{d\gamma_i}{dT} \right) \mathbf{H}. \quad (9)$$

Moreover as shown by Subramanian (1985) this expression for the thermocapillary velocity of a single bubble also applies when  $T_\infty$  is any harmonic function having singularities outside the space occupied by the bubble, provided that  $\mathbf{H}$  is replaced by  $(\nabla T_\infty)_0$ , i.e. the ambient temperature gradient evaluated at the centre of the bubble.

Consider next  $N$  bubbles whose parameters  $a_i$  and  $d\gamma_i/dT$  satisfy the relations

$$a_1 \left( -\frac{d\gamma_1}{dT} \right) = a_2 \left( -\frac{d\gamma_2}{dT} \right) = \dots = a_N \left( -\frac{d\gamma_N}{dT} \right) \equiv -2\mu b, \quad (10)$$

where  $b$  is a constant, so that

$$\mathbf{U}_1^{(0)} = \mathbf{U}_2^{(0)} = \dots = \mathbf{U}_N^{(0)}.$$

Now, if the ambient temperature and velocity fields,  $T_\infty$  and  $\mathbf{u}_\infty$  respectively, satisfy the relation

$$\mathbf{u}_\infty = b\nabla T_\infty, \quad (11)$$

then the flow field remains irrotational and is given by

$$\mathbf{u} = b\nabla T \quad \text{with} \quad p = 0, \quad (12)$$

and all the bubbles will be stationary, i.e.

$$\mathbf{U}_i = 0. \quad (13)$$

The relations (12) and (13) can be easily verified by direct substitution into (4)–(8) and using the conditions (3) and (10).

A physical interpretation of this result can be obtained by first considering a single bubble immersed in the ambient fields  $T_\infty, \mathbf{u}_\infty$  which satisfy (11). It can easily be verified then that the resulting flow field  $\mathbf{u}' = b\nabla T'$ , where  $T'$  is the new temperature profile, is that formed by a stationary bubble in an irrotational inviscid flow which also satisfies the shear stress balance equation (8) as well as the conditions of zero force. Now another bubble is placed in the fields  $T'$  and  $\mathbf{u}'$ , which in turn can be viewed as the ambient fields. The same argument can be applied again except for the generation

of further reflections by the presence of the first bubble, which, once again, satisfy (11). Finally, using the same argument repeatedly, we conclude that if any number of bubbles are placed in the fields  $T_\infty, \mathbf{u}_\infty$  the results (12) and (13) are valid, showing that, as in the case of an isolated bubble, the effects of the temperature and of the flow field on the velocity of the bubbles exactly balance each other. Although (10) is of course totally unphysical, the resulting analysis just described will prove useful later on in constructing the solution to our mathematical problem.

The thermocapillary velocities of  $N$  identical bubbles immersed in a quiescent ambient fluid with constant temperature gradient  $\nabla T_\infty = \mathbf{H}$  can be easily obtained using (11) and (13). In fact, by superimposing the uniform flow fields  $\mathbf{u}_\infty = b\mathbf{H}$  and  $\mathbf{u}_\infty = -b\mathbf{H}$ , we see that the former will 'balance' the effect of the temperature gradient (cf. (13)), while the latter will produce a net uniform velocity. Finally, we may conclude that each bubble will move with the same constant velocity  $U^{(0)} = -b\mathbf{H}$  as if it were alone, in agreement with the result obtained by Acrivos *et al.* (1990).

Another interesting application of (12) and (13), which will be found useful when performing the analysis of §5.1, pertains to the motion of a bubble near a stress-free plane on which a fixed temperature gradient  $\nabla T = \mathbf{H}$  is imposed perpendicular to the plane on the side facing the bubble. By the method of images, the plane can then be replaced by an identical second bubble placed at the position symmetric about the plane and with  $\nabla T = -\mathbf{H}$  on the other side facing the second bubble, so that the stress-free condition on the plane is satisfied identically. Now, repeating the same argument as before, i.e. by superimposing the two flows  $\mathbf{u}_\infty = \pm b\nabla T_\infty$  and noting that the flow  $\mathbf{u}_\infty = b\nabla T_\infty$  balances the effect of the temperature gradient according to (11)–(13), we conclude that the original bubble will move with a constant velocity  $-b\mathbf{H}$  as if the plane were absent.

### 3. The solution of the two-bubble problem

In this section we shall study the motion of two bubbles in an unbounded fluid resulting from the presence of a constant ambient temperature gradient  $\mathbf{H}$ . The solution of this problem is required in §4 to determine the average velocity of a bubble immersed in a suspension of bubbles of a different size.

As noted earlier, the motion of two unequal-size bubbles can be determined by first solving the thermal problem to find the temperature distribution and the surface tension distribution on the surface of the bubbles, and then solving the hydrodynamic problem to find the velocity of the bubbles. This problem is greatly simplified when the two bubbles are aligned with the temperature gradient (i.e. with axisymmetric geometry), so that spherical bipolar coordinates can be used, as shown by Meyyappan, Wilcox & Subramanian (1983) and by Keh & Chen (1990). For the general case of two bubbles with radii  $a_1$  and  $a_2 \equiv \lambda a_1$  and arbitrary orientation, Anderson (1985) applied the direct reflection method to determine the bubble velocities up to terms of  $O(R^{-6})$ , with  $R \equiv r/a_1$ , where  $r$  denotes the centre-to-centre distance between the two bubbles, while Keh & Chen (1992, 1993) used a boundary collocation technique to find the mobility functions of two drops and two bubbles for  $\lambda = \frac{1}{2}, 1$  and  $2$ .

In this section, we shall show that higher accuracy can be reached by applying the method of twin multipole expansions, an approach that was also followed in a recent work by Satrape (1992). In both cases the solution is expanded as an infinite series whose coefficients are to be determined by satisfying the boundary conditions of the problem. But whereas in the approach adopted by Satrape the series expansion was truncated and these coefficients were determined to the desired accuracy for every value

of  $R$  and  $\lambda$ , in what follows the coefficients of the multipole expansions will be expanded as power series in  $\lambda/R$  and  $1/R$  and therefore expansions will be derived for the bubble velocities in a form where their dependence on  $R$  and  $\lambda$  is factored out. Consequently, although our results are equivalent to those obtained by Satrape (1992), they are expressed in a more convenient form for the purpose of obtaining the ensemble-averaged velocity of a test bubble in the suspension.

### 3.1. The general solution

Owing to the linearity of the thermal and the hydrodynamic problems, the general case of arbitrary orientation of the two bubbles, with respect to the ambient temperature gradient  $\mathbf{H}$  is decomposed into two problems, in which the centreline  $\mathbf{r}$  is parallel and perpendicular to  $\mathbf{H}$ , respectively; the former case is obviously axisymmetric and the latter is not.

Following Jeffrey & Onishi (1984), two sets of spherical polar coordinates  $(\rho_\alpha, \theta_\alpha, \phi)$  are chosen ( $\alpha = 1, 2$ ) to describe the two-bubble geometry.

First consider the thermal problem, which is a special case of a more general problem solved by Thovert & Acrivos (1989) involving two spheres of different sizes and equal but arbitrary heat conductivities embedded in an ambient temperature field of constant gradient.

The temperature distribution outside the bubbles is expanded, for the parallel case ( $m = 0$ ) and the perpendicular case ( $m = 1$ ) respectively, as

$$T_m = (-1)^{m(\alpha+1)} H \rho_\alpha Y_{m1}(\theta_\alpha, \phi) + H \sum_{n=m}^{\infty} \left[ g_{mn}^{(\alpha)} \left( \frac{a_\alpha}{\rho_\alpha} \right)^{n+1} Y_{mn}(\theta_\alpha, \phi) + g_{mn}^{(3-\alpha)} \left( \frac{a_{3-\alpha}}{\rho_{3-\alpha}} \right)^{n+1} Y_{mn}(\theta_{3-\alpha}, \phi) \right], \quad (14)$$

with  $H = |\mathbf{H}|$ , where  $Y_{mn}(\theta_\alpha, \phi)$  are spherical surface harmonics, while the coefficients  $g_{mn}^{(\alpha)}$ , which depend on  $\lambda$  and  $R$ , are to be determined from the boundary conditions. Expanding  $g_{mn}^{(\alpha)}$  as a double power series in terms of  $t_\alpha \equiv a_\alpha/r$ ,

$$g_{mn}^{(\alpha)} = (-1)^{(m+1)\alpha} a_\alpha \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} G_{n p q}^m t_\alpha^p t_{3-\alpha}^q, \quad (15)$$

and substituting (14) and (15) into (3), yields the recurrence relation:

$$G_{n p q}^m = (-1)^{(m+1)} \binom{n}{n+1} \sum_{s=m}^{\infty} \binom{n+s}{n+m} G_{s, q-s-2, p-n+1}^m, \quad (16)$$

with

$$G_{100}^m = \frac{1}{2} (-1)^{(m+1)}. \quad (17)$$

Equations (16) and (17) completely determine the temperature field.

We next turn to the hydrodynamic problem. The pressure and flow fields can be written as the sum of the contributions of singularities at the centre of the bubbles (Jeffrey & Onishi 1984):

$$p = p^{(\alpha)} + p^{(3-\alpha)},$$

and

$$\mathbf{u} = \mathbf{u}^{(\alpha)} + \mathbf{u}^{(3-\alpha)}, \quad (18)$$

where

$$p^{(\alpha)} = \mu \sum_{m=0}^{\infty} \sum_{n=m}^{\infty} \frac{1}{a_\alpha} p_{mn}^{(\alpha)} \left( \frac{a_\alpha}{\rho_\alpha} \right)^{n+1} Y_{mn}(\theta_\alpha, \phi), \quad (19)$$

and

$$\begin{aligned} \mathbf{u}^{(\alpha)} = & \sum_{m=0}^{\infty} \sum_{n=m}^{\infty} \left\{ \nabla \times \left[ \rho_{\alpha} q_{mn}^{(\alpha)} \left( \frac{a_{\alpha}}{\rho_{\alpha}} \right)^{n+1} Y_{mn}(\theta_{\alpha}, \phi) \right] + a_{\alpha} \nabla \left[ v_{mn}^{(\alpha)} \left( \frac{a_{\alpha}}{\rho_{\alpha}} \right)^{n+1} Y_{mn}(\theta_{\alpha}, \phi) \right] \right. \\ & \left. - \frac{n-2}{2n(2n-1)a_{\alpha}} \rho_{\alpha}^2 \nabla \left[ p_{mn}^{(\alpha)} \left( \frac{a_{\alpha}}{\rho_{\alpha}} \right)^{n+1} Y_{mn}(\theta_{\alpha}, \phi) \right] + \frac{n+1}{n(2n-1)a_{\alpha}} \rho_{\alpha} p_{mn}^{(\alpha)} \left( \frac{a_{\alpha}}{\rho_{\alpha}} \right)^{n+1} Y_{mn}(\theta_{\alpha}, \phi) \right\}. \end{aligned} \quad (20)$$

The coefficients  $p_{mn}^{(\alpha)}$ ,  $v_{mn}^{(\alpha)}$  and  $q_{mn}^{(\alpha)}$  are functions only of  $r$  and  $\lambda$ , and are to be determined from the boundary conditions.

To simplify the application of the boundary conditions, we follow Happel & Brenner (1965) and Jeffrey & Onishi (1984) in first constructing the following three scalar equations:

$$\hat{\rho}_{\alpha} \cdot \mathbf{u} = \hat{\rho}_{\alpha} \cdot \mathbf{U}_{\alpha} = \sum_{m=0}^{\infty} \sum_{n=m}^{\infty} \chi_{mn}^{(\alpha)} Y_{mn}(\theta_{\alpha}, \phi), \quad (21)$$

$$\frac{1}{\mu} \left[ a_{\alpha}^2 \nabla \cdot (\boldsymbol{\sigma} \cdot \hat{\rho}_{\alpha}) - \frac{\partial}{\partial \rho_{\alpha}} (\rho_{\alpha} \cdot \boldsymbol{\sigma} \cdot \rho_{\alpha}) \right] = \frac{a_{\alpha}^2}{\mu} \nabla_s \cdot \mathbf{f}_s = \sum_{m=0}^{\infty} \sum_{n=m}^{\infty} \psi_{mn}^{(\alpha)} Y_{mn}(\theta_{\alpha}, \phi), \quad (22)$$

$$\frac{a_{\alpha}}{\mu} \rho_{\alpha} \cdot [\nabla \times (\boldsymbol{\sigma} \cdot \hat{\rho}_{\alpha})] = \frac{a_{\alpha}}{\mu} \rho_{\alpha} \cdot (\nabla_s \times \mathbf{f}_s) = \sum_{m=0}^{\infty} \sum_{n=m}^{\infty} \omega_{mn}^{(\alpha)} Y_{mn}(\theta_{\alpha}, \phi), \quad (23)$$

where  $\hat{\rho}_{\alpha} = \rho_{\alpha} / \rho_{\alpha}$ ,  $\mathbf{f}_s = (\mathbf{I} - \hat{\rho}_{\alpha} \hat{\rho}_{\alpha}) \cdot (\boldsymbol{\sigma} \cdot \hat{\rho}_{\alpha})$  and  $\nabla_s = (\mathbf{I} - \hat{\rho}_{\alpha} \hat{\rho}_{\alpha}) \cdot \nabla$ , while the functions  $\chi_{mn}^{(\alpha)}$ ,  $\psi_{mn}^{(\alpha)}$  and  $\omega_{mn}^{(\alpha)}$  can be obtained from the boundary conditions (7) and (8).

Next, we use the scalar equations (21)–(23) just constructed to find the relations for the coefficients  $p_{mn}^{(\alpha)}$ ,  $v_{mn}^{(\alpha)}$  and  $q_{mn}^{(\alpha)}$  in (19) and (20).

Substituting (18) into the boundary conditions (21)–(23), expressing all the functions of  $\rho_{3-\alpha}$ ,  $\theta_{3-\alpha}$  and  $\phi$  in terms of  $r$ ,  $\rho_{\alpha}$ ,  $\theta_{\alpha}$  and  $\phi$  by the transformation rule (see equation (2.1) in Jeffrey & Onishi 1984) and then using the orthogonality relations of  $Y_{mn}(\theta_{\alpha}, \phi)$  yields three relations for  $p_{mn}^{(\alpha)}$ ,  $v_{mn}^{(\alpha)}$  and  $q_{mn}^{(\alpha)}$ . For convenience in later computations, the first two relations are reorganized. Finally, we obtain the following three general recurrence relations for  $p_{mn}^{(\alpha)}$ ,  $v_{mn}^{(\alpha)}$  and  $q_{mn}^{(\alpha)}$ :

$$2(2n+1) \left\{ (n+1) v_{mn}^{(\alpha)} - \frac{n}{2(2n+3)} \sum_{s=m}^{\infty} \binom{n+s}{n+m} t_{\alpha}^{n-1} t_{3-\alpha}^s p_{ms}^{(3-\alpha)} \right\} = \psi_{mn}^{(\alpha)} + 2(n^2-1) \chi_{mn}^{(\alpha)}, \quad (24)$$

$$\begin{aligned} 2(2n+1) \left\{ \frac{(n+1)}{2(2n-1)} p_{mn}^{(\alpha)} + \sum_{s=m}^{\infty} \binom{n+s}{n+m} t_{\alpha}^{n-1} t_{3-\alpha}^s \left[ im(-1)^{\alpha} q_{ms}^{(3-\alpha)} + nv_{ms}^{(3-\alpha)} t_{3-\alpha}^2 \right. \right. \\ \left. \left. + \left( \frac{(n^2-m^2)(sn-2s-2n+1)}{s(2s-1)(2n-1)(n+s)} - \frac{n(s-2)}{2s(2s-1)} \right) p_{ms}^{(3-\alpha)} \right] \right\} = \psi_{mn}^{(\alpha)} + 2n(n+2) \chi_{mn}^{(\alpha)}, \end{aligned} \quad (25)$$

$$n(n+1)(n+2) q_{mn}^{(\alpha)} + (n-1) \sum_{s=m}^{\infty} \binom{n+s}{n+m} t_{\alpha}^n t_{3-\alpha}^s \left[ ns q_{ms}^{(3-\alpha)} t_{3-\alpha} + (-1)^{3-\alpha} \frac{m}{s} i p_{ms}^{(3-\alpha)} \right] = \omega_{mn}^{(\alpha)}. \quad (26)$$

The force-free condition on the bubbles yields

$$p_{01}^{(\alpha)} = p_{11}^{(\alpha)} = 0,$$

which, together with (24)–(26) completely determine the coefficients  $p_{mn}^{(\alpha)}$ ,  $v_{mn}^{(\alpha)}$ ,  $q_{mn}^{(\alpha)}$  and  $U_{\alpha}$ , once  $\chi_{mn}^{(\alpha)}$ ,  $\psi_{mn}^{(\alpha)}$  and  $\omega_{mn}^{(\alpha)}$  have been calculated from (7), (8), (21)–(23).

Before applying these general results to the parallel and the perpendicular cases separately, we define the mobility functions, which are the quantities of most interest.

The mobility functions  $A_{\alpha\beta}$  (for the axisymmetric case) and  $B_{\alpha\beta}$  (for the perpendicular case) ( $\alpha, \beta = 1, 2$ ) are defined through the relations

$$U_\alpha = \mathbf{M}_{\alpha\alpha} \cdot U_\alpha^{(0)} + \mathbf{M}_{\alpha(3-\alpha)} \cdot U_{(3-\alpha)}^{(0)}, \quad (27)$$

for the velocities of two bubbles arbitrarily oriented relative to  $\mathbf{H}$ , where

$$\mathbf{M}_{\alpha\beta} = A_{\alpha\beta}(r, \lambda) \frac{\mathbf{r}\mathbf{r}}{r^2} + B_{\alpha\beta}(r, \lambda) \left( \mathbf{I} - \frac{\mathbf{r}\mathbf{r}}{r^2} \right). \quad (28)$$

In this definition, it is implicitly assumed that, in both the parallel and perpendicular cases, the velocity of each bubble is parallel to  $\mathbf{H}$  as can be easily inferred from the possible general form  $U_\alpha = [f_1(\lambda, r)\mathbf{I} + f_2(\lambda, r)\mathbf{r}\mathbf{r}] \cdot \mathbf{H}$  and from the symmetry of the problem.

Finally, it is easy to see from the result of §2 that if  $U_1^{(0)} = U_2^{(0)}$  then  $U_1 = U_2 = U_1^{(0)} = U_2^{(0)}$ . Therefore (27) and (28) give

$$A_{\alpha\alpha}(r, \lambda) + A_{\alpha(3-\alpha)}(r, \lambda) = 1, \quad (29)$$

$$B_{\alpha\alpha}(r, \lambda) + B_{\alpha(3-\alpha)}(r, \lambda) = 1. \quad (30)$$

It is also easy to see by interchanging the labels 1 and 2, that

$$A_{\alpha\beta}(r, \lambda) = A_{(3-\alpha)(3-\beta)}(r, \lambda^{-1}), \quad (31)$$

$$B_{\alpha\beta}(r, \lambda) = B_{(3-\alpha)(3-\beta)}(r, \lambda^{-1}). \quad (32)$$

So, our problem reduces to finding two independent scalar mobility functions, say,  $A_{11}$  and  $B_{11}$ .

### 3.2. Determination of $A_{\alpha\beta}$ (i.e. the parallel case)

Consider first the axisymmetric case where the ambient temperature gradient  $\mathbf{H}$  is parallel to the centreline  $\mathbf{r}$ . Since  $U_1^{(0)}$  and  $U_2^{(0)}$  are also parallel to  $\mathbf{H}$  and therefore to  $\mathbf{r}$ , (27) and (28) yield

$$U_1 = A_{11} U_1^{(0)} + A_{12} U_2^{(0)}, \quad (33)$$

where  $A_{12} = 1 - A_{11}$  (cf. (29)),  $U_1 = |U_1|$  and similarly for  $U_1^{(0)}$  and  $U_2^{(0)}$ . In turn,  $A_{11}$  can be determined by considering two separate cases in which the velocities of the two bubbles are either parallel or antiparallel to each other, i.e.  $U_1^{(0)} = \pm U_2^{(0)} = U_0$ . Denoting by  $U_+^{(1)}$  and  $U_-^{(1)}$  the resulting velocities of bubble 1, we find that  $A_{11} = (U_+^{(1)} + U_-^{(1)})/2U_0$ , where  $U^{(1)}$  (the  $\pm$  subscript is hereby omitted for simplicity) can be determined through (14)–(20), subject to the following boundary conditions (cf. (21)–(23)):

$$\psi_{0n}^{(\alpha)} = -6(\pm 1)^\alpha (n+1) \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} G_{n\,pq}^m t_\alpha^p t_{3-\alpha}^q \equiv U_0 \sum_{p=1}^{\infty} \sum_{q=0}^{\infty} B_{n\,pq} t_\alpha^p t_{3-\alpha}^q, \quad (34)$$

$$\chi_{0n}^{(\alpha)} = -(\mp 1)^{3-\alpha} \delta_{n1} U^{(\alpha)}, \quad (35)$$

$$\omega_{0n}^{(\alpha)} = 0, \quad (36)$$

where the  $\pm$  sign refers to the parallel and anti-parallel cases, respectively.

Now, we expand  $p_{0n}^{(\alpha)}$ ,  $q_{0n}^{(\alpha)}$  and  $v_{0n}^{(\alpha)}$  in (19) and (20):

$$p_{0n}^{(\alpha)} = (\mp 1)^{3-\alpha} U_0 \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} P_{npq} t_{\alpha}^p t_{3-\alpha}^q, \quad (37)$$

$$q_{0n}^{(\alpha)} = (\mp 1)^{3-\alpha} U_0 \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} Q_{npq} t_{\alpha}^p t_{3-\alpha}^q, \quad (38)$$

$$v_{0n}^{(\alpha)} = (\mp 1)^{3-\alpha} U_0 \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} \frac{1}{2n+1} V_{npq} t_{\alpha}^p t_{3-\alpha}^q, \quad (39)$$

where, from the force-free condition on the bubbles,

$$P_{1pq} = 0. \quad (40)$$

Note that in this (i.e. the axisymmetric) case,  $q_{0n}^{(\alpha)}$ , the coefficients of the azimuthal term in the velocity field (20), are identically zero on account of the symmetry of the problem. Moreover the velocities of the bubbles, defined following Jeffrey & Onishi (1984) as  $U_1 = U^{(1)}\mathbf{H}/H$  and  $U_2 = \pm U^{(2)}\mathbf{H}/H$  again can be expanded as

$$U^{(\alpha)} = (\mp 1)^{3-\alpha} U_0 \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} U_{pq} t_{\alpha}^p t_{3-\alpha}^q. \quad (41)$$

Substituting (37)–(41) into the general recurrence relations (24)–(26) we obtain, for  $n \geq 2$ ,

$$P_{npq} = \frac{2(2n-1)}{n+1} \left\{ -\frac{B_{npq}}{2(2n+1)} \mp \sum_{s=1}^{\infty} \binom{n+s}{n} \left[ \frac{n}{2s+1} V_{s, q-s-2, p-n+1} + \left( \frac{n^2(sn-2s-2n+1)}{s(2s-1)(2n-1)(n+s)} - \frac{n(s-2)}{2s(2s-1)} \right) P_{s, q-s, p-n+1} \right] \right\}, \quad (42)$$

$$V_{npq} = \frac{2n+1}{n+1} \left\{ -\frac{B_{npq}}{2(2n+1)} \pm \frac{n}{2(2n+3)} \sum_{s=1}^{\infty} \binom{n+s}{n} P_{s, q-s, p-n+1} \right\}, \quad (43)$$

and for  $n=1$ ,

$$U_{pq} = \frac{1}{6} B_{1pq} + \left\{ P_{1pq} \pm \sum_{s=1}^{\infty} \binom{1+s}{1} \left[ \frac{1}{2s+1} V_{s, q-s-2, p} + \frac{1-s}{2s(2s-1)} P_{s, q-s, p} \right] \right\}. \quad (44)$$

Finally, the values of  $U_+^{(1)}$  and  $U_-^{(1)}$  can be evaluated through (41)–(44). In agreement with the analysis preceding equation (13), we find that  $U_+^{(1)} = U_0$ , and on substituting the value of  $U^{(1)}$  into  $A_{11} = \frac{1}{2}(1 + U^{(1)}/U_0)$  we find that

$$A_{11} = \frac{1}{2} + \frac{1}{2} \sum_{k=0}^{\infty} \sum_{q=0}^k U_{(k-q)q}^- \frac{a_1^{k-q} a_2^q}{r^k} = \sum_{k=0}^{\infty} \frac{A_{11}^{(k)}(\lambda)}{R^k}, \quad (45)$$

with  $R = r/a_1$ , where

$$A_{11}^{(k)}(\lambda) = \frac{1}{2} \delta_{k1} + \frac{1}{2} \sum_{q=0}^k U_{(k-q)q}^- \lambda^q. \quad (46)$$

Here  $U_{pq}^-$  is given by (44) for the anti-parallel case. The first few terms of (45) are

$$A_{11}(R, \lambda) = 1 - \frac{\lambda^3}{R^3} - 2 \frac{\lambda^3}{R^6} - 3 \frac{\lambda^5}{R^8} - 4 \frac{\lambda^6}{R^9} - 4 \frac{\lambda^7}{R^{10}} - \frac{9\lambda^6 + 6\lambda^8}{R^{11}} - \frac{8\lambda^6 + 5\lambda^9}{R^{12}} + O\left(\frac{1}{R^{13}}\right). \quad (47)$$

This extends Anderson's (1985) result, which consists of the first three terms of (47).



$R-1-\lambda$	$\lambda = \frac{1}{20}$	$\lambda = \frac{1}{10}$	$\lambda = \frac{1}{2}$	$\lambda = 1$	$\lambda = 2$	$\lambda = 10$	$\lambda = 20$
0	0.99962	0.99734	0.89534	0.71979	0.49589	0.16307	0.09714
0.01	0.99967	0.99770	0.90606	0.74049	0.51759	0.16822	0.09930
0.05	0.99976	0.99834	0.92943	0.79161	0.57960	0.18719	0.10764
0.10	0.99981	0.99871	0.94385	0.82645	0.62842	0.20791	0.11744
0.50	0.99995	0.99964	0.97971	0.92262	0.78802	0.31893	0.18058
1.00	0.99998	0.99987	0.99088	0.95932	0.86696	0.41065	0.24150
2.00	1.00000	0.99996	0.99694	0.98382	0.93447	0.54142	0.33982

TABLE 1. The function  $A_{11}(R, \lambda)$  obtained by summing the series (45) to terms  $O(1/R^{120})$ ,  
 $R = r/a_1$ ,  $\lambda = a_2/a_1$

Some typical numerical values of  $A_{11}(R, \lambda)$  obtained by summing the series (45) up to terms of  $O(1/R^{120})$ , are given in table 1.

### 3.3. Determination of $B_{\alpha\beta}$ (i.e. the perpendicular case)

When  $H$  is perpendicular to the centreline  $r$ , (27) yields

$$U_1 = B_{11} U_1^{(0)} + B_{12} U_2^{(0)}, \quad (48)$$

where  $B_{12} = 1 - B_{11}$  (cf. (30)).

Next we proceed as in the previous case, with  $m = 1$  replacing  $m = 0$  in (34)–(40), substituting (37)–(41) into the general recurrence relations (24)–(26) yields, for  $n \geq 2$ ,

$$P_{npq} = \frac{2(2n-1)}{n+1} \left\{ -\frac{B_{npq}}{2(2n+1)} \mp \sum_{s=1}^{\infty} \binom{n+s}{n+1} \left[ Q_{s, q-s-1, p-n+1} + \frac{n}{2s+1} V_{s, q-s-2, p-n+1} \right. \right. \\ \left. \left. + \left( \frac{(n^2-1)(sn-2s-2n+1)}{s(2s-1)(2n-1)(n+s)} - \frac{n(s-2)}{2s(2s-1)} \right) P_{s, q-s, p-n+1} \right] \right\}, \quad (49)$$

$$V_{npq} = \frac{2n+1}{n+1} \left\{ -\frac{B_{npq}}{2(2n+1)} \pm \frac{n}{2(2n+3)} \sum_{s=1}^{\infty} \binom{n+s}{n+1} P_{s, q-s, p-n+1} \right\}, \quad (50)$$

$$Q_{npq} = \pm \frac{n-1}{n(n+1)(n+2)(n+3)} \sum_{s=1}^{\infty} \binom{n+s}{n+1} \left[ ns Q_{s, q-s-1, p-n} + \frac{1}{s} P_{s, q-s, p-n} \right], \quad (51)$$

and for  $n = 1$ ,

$$U_{pq} = \frac{1}{6} B_{1pq} + \left\{ P_{1pq} \pm \sum_{s=1}^{\infty} \binom{1+s}{2} \left[ \frac{1}{2s+1} V_{s, q-s-2, p-n+1} + \frac{2-s}{2s(2s-1)} P_{s, q-s, p} \right] \right\}. \quad (52)$$

Finally, the bubble velocity  $U_\alpha$  is evaluated by substituting (52) into (41). For the case  $U_1^{(0)} = U_2^{(0)}$ , we find  $U_1 = U_2 = U_0$ , in agreement with (13), and combining the results of the two cases  $U_1^{(0)} = \pm U_2^{(0)}$ , we obtain

$$B_{11} = \frac{1}{2} + \frac{1}{2} \sum_{k=0}^{\infty} \sum_{q=0}^k U_{(k-q)q}^- \frac{a_1^{k-q} a_2^q}{r^k} = \sum_{k=0}^{\infty} \frac{B_{11}^{(k)}(\lambda)}{R^k}, \quad (53)$$

where

$$B_{11}^{(k)}(\lambda) = \frac{1}{2} \delta_{k1} + \frac{1}{2} \sum_{q=0}^k U_{(k-q)q}^- \lambda^q. \quad (54)$$

The first few terms of (53) are

$$B_{11}(R, \lambda) = 1 + \frac{\lambda^3}{2R^3} + \frac{\lambda^3}{4R^6} + \frac{\lambda^5}{4R^8} + \frac{\lambda^6}{8R^9} + \frac{\lambda^7}{4R^{10}} + \frac{4\lambda^6 + \lambda^8}{8R^{11}} + \frac{\lambda^6 + 4\lambda^9}{16R^{12}} + O\left(\frac{1}{R^{13}}\right). \quad (55)$$

$R-1-\lambda$	$\lambda = \frac{1}{20}$	$\lambda = \frac{1}{10}$	$\lambda = \frac{1}{2}$	$\lambda = 1$	$\lambda = 2$	$\lambda = 10$	$\lambda = 20$
0	1.00008	1.00055	1.02281	1.07038	1.56443	1.37727	1.43240
0.01	1.00008	1.00052	1.02214	1.06900	1.15459	1.37620	1.43177
0.05	1.00007	1.00044	1.01989	1.06408	1.14768	1.37201	1.42929
0.10	1.00005	1.00038	1.01766	1.05884	1.13986	1.36688	1.42621
0.50	1.00002	1.00014	1.00835	1.03332	1.09521	1.32943	1.40272
1.00	1.00001	1.00006	1.00413	1.01892	1.06321	1.28973	1.37581
2.00	1.00000	1.00002	1.00148	1.00788	1.03216	1.22774	1.32883

TABLE 2. The function  $B_{11}(R, \lambda)$  obtained by summing the series (53) to terms  $O(1/R^{120})$ ,  
 $R = r/a_1$ ,  $\lambda = a_2/a_1$

Again this generalizes Anderson's (1985) result, which consists of the first three terms of (55).

Some typical numerical values of  $B_{11}(R, \lambda)$  obtained by summing the series (53) up to terms of  $O(1/R^{120})$ , are given in table 2. Also we note that our numerical values for the velocity of the bubbles and those given by Satrape (1992) were found to agree to five significant figures.

## 5. The average velocity of a bubble

Acrivos *et al.* (1990) calculated the ensemble-averaged bubble velocity in a monodisperse suspension of bubbles. In this section, we extend the calculation to bidisperse dilute suspensions by determining the average velocity of bubble 1 in a space-filling suspension of bubbles 2.

As was pointed out by Acrivos *et al.* (1990), such a calculation requires the application of a renormalization procedure which takes into account the following two constraints: (i) that the ensemble-averaged velocity at any point in the suspension is zero and (ii) that the corresponding ensemble-averaged temperature gradient equals the imposed temperature gradient  $H$ . In terms of ensemble averages, these constraints for a dilute suspension of bubbles 2 are

$$\int \mathbf{u}P(\mathbf{r})d\mathbf{r} = 0 \quad \text{and} \quad \int (\nabla T - H)P(\mathbf{r})d\mathbf{r} = 0, \quad (56)$$

where  $P(\mathbf{r})$  is the unconditional probability that the centre of a single bubble 2 is at  $\mathbf{r}$ , while  $\mathbf{u} = \mathbf{u}(\mathbf{r})$  and  $\nabla T = \nabla T(\mathbf{r})$  are, respectively, the velocity and the temperature gradient at the origin, when a single bubble 2 is present with its centre at  $\mathbf{r}$ . Here, we have used the assumption that the suspension is so dilute that the probability of two bubbles 2 being at distance  $r \sim O(a_1 + a_2)$  from the origin is of  $O(c_2^2)$  and can therefore be neglected, where  $c_2$  is the volume fraction occupied by bubbles 2.

Next, consider the average velocity of a test bubble 1 with its centre at the origin,

$$\bar{U}_1 = U_1^{(0)} + \int (U_1 - U_1^{(0)})P(\mathbf{r}|\mathbf{0})d\mathbf{r}, \quad (57)$$

where  $U_1 = U_1(\mathbf{0}|\mathbf{r})$  is the velocity of a bubble 1 at the origin in the presence of a single bubble 2 with its centre at  $\mathbf{r}$  and  $P(\mathbf{r}|\mathbf{0})$  is the conditional probability of having a single bubble 2 at  $\mathbf{r}$  given that a bubble 1 is at the origin.

All the integrals in (56) and (57) are non-convergent, with the divergent terms representing the zeroth-order reflections of the velocity and temperature fields, i.e. the velocity of bubble 1 at the origin induced by the velocity and temperature disturbance

of a single bubble 2 at  $\mathbf{r}$ . Therefore, as shown by Acrivos *et al.* (1990), these identical divergent terms can be subtracted from each other, and  $\bar{U}_1$  can be determined by evaluating the remaining higher-order terms representing higher-order reflections.

Thus we arrive at

$$\bar{U}_1 = U_1^{(0)} + \int \mathbf{u}(\mathbf{r})[P(\mathbf{r}|\mathbf{0}) - P(\mathbf{r})] d\mathbf{r} + \frac{U_1^{(0)}}{H} \int (\nabla T - H)[P(\mathbf{r}|\mathbf{0}) - P(\mathbf{r})] d\mathbf{r} + \int W(\mathbf{r}) P(\mathbf{r}|\mathbf{0}) d\mathbf{r}, \quad (58)$$

where  $W(\mathbf{r})$ , defined by

$$U_1 = U_1^{(0)} + \mathbf{u} + \frac{U_1^{(0)}}{H} (\nabla T - H) + W, \quad (59)$$

is obtained from (27) by retaining only the terms of order higher than  $O(1/R^3)$  in (47) and (55).

For well-mixed suspensions of bubbles 2, the probability and conditional probability functions are given by (Jeffrey 1973)

$$P(\mathbf{r}) = \frac{c_2}{\frac{4}{3}\pi a_2^3} \quad (60)$$

and

$$P(\mathbf{r}|\mathbf{0}) = 0 \quad \text{for } r < a_1 + a_2, \quad (61)$$

$$P(\mathbf{r}|\mathbf{0}) = P(\mathbf{r}) = \frac{c_2}{\frac{4}{3}\pi a_2^3} \quad \text{for } r \geq a_1 + a_2,$$

where we have assumed that, in the presence of the test bubble 1, the distribution of bubbles 2 is still uniform outside the exclusion layer  $r = a_1 + a_2$ . Substituting the probability distributions (60) and (61) into the first two integrals in (58) yields

$$\bar{U}_1 = U_1^{(0)} - c_2 U_2^{(0)} - \frac{1}{2} c_2 U_1^{(0)} + c_2 \int_{r \geq a_1 + a_2} W(\mathbf{r}) \frac{d\mathbf{r}}{\frac{4}{3}\pi a_2^3}. \quad (62)$$

Now, since in any physically relevant situation the surface tensions of bubbles 1 and 2 have the same temperature dependence, i.e.  $d\gamma_1/dT = d\gamma_2/dT$ , so that  $U_2^{(0)} = \lambda U_1^{(0)}$ , (62) reduces to

$$\bar{U}_1 = \bar{U}_1^{(0)} \{1 - \frac{3}{2} c_1 - S(\lambda) c_2 + \dots\}, \quad (63)$$

where the dependence of  $\bar{U}_1$  on  $c_1$  is given by Acrivos *et al.* (1990). In the above, terms of  $O(c_1^2)$ ,  $O(c_2^2)$ ,  $O(c_1 c_2)$  and higher have been neglected since the analysis is restricted to dilute suspensions. Also

$$S(\lambda) \equiv \lambda + \frac{1}{2} + I(\lambda), \quad (64)$$

with

$$I(\lambda) = \frac{\lambda - 1}{\lambda^3} \int_{1+\lambda}^{\infty} (A_{11} + 2B_{11} - 3) R^2 dR. \quad (65)$$

On substituting (45) and (53) into (64) and (65) we then obtain

$$S(\lambda) = \lambda + \frac{1}{2} + (\lambda - 1) \frac{(1 + \lambda)^3}{\lambda^3} \sum_{n=6}^{\infty} \frac{A_{11}^{(n)} + 2B_{11}^{(n)}}{(n-3)(1+\lambda)^n}, \quad (66)$$

where  $A_{11}^{(n)}$  and  $B_{11}^{(n)}$  are given by (46) and (54). The function  $S(\lambda)$  is tabulated in table 3 and is plotted in figure 1 together with its asymptotic expressions to be derived below

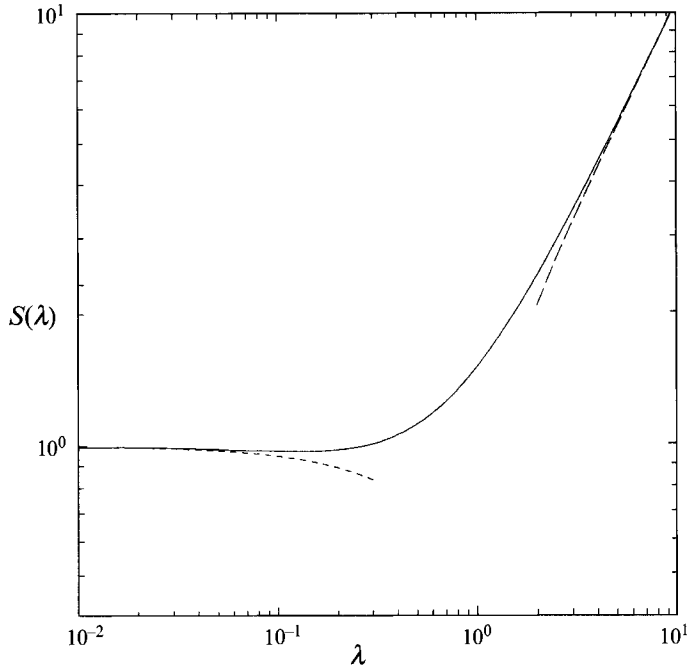


FIGURE 1. The function  $S(\lambda)$  with the solid line representing the computed results by summing the series in (66), while the dashed lines on the left and right sides are based on the asymptotic expressions (84), for  $\lambda \rightarrow 0$  and (76), for  $\lambda \rightarrow \infty$ , respectively.

$\lambda$	$S(\lambda)$	From (84)	From (76)	From (67)
$\frac{1}{64}$	0.9922	0.9911	—	0.9854
$\frac{1}{32}$	0.9858	0.9822	—	0.9729
$\frac{1}{16}$	0.9744	0.9644	—	0.9533
$\frac{1}{8}$	0.9663	0.9288	—	0.9323
$\frac{1}{4}$	0.9865	0.8575	—	0.9420
$\frac{1}{2}$	1.1112	0.7150	—	1.0741
1	1.5000	0.4300	0.6600	1.5000
2	2.4372	—	2.0800	2.4815
4	4.4257	—	4.2900	4.4880
8	8.4415	—	8.3950	8.4952
16	16.4620	—	16.4475	16.4985
32	32.4778	—	32.4737	32.4996
64	64.4870	—	64.4869	64.4999

TABLE 3. Values of the function  $S(\lambda)$  computed by summing the series in (66) to terms  $O(1/R^{120})$  and then extrapolating the result, as well as from the corresponding asymptotic expressions for  $\lambda \ll 1$  and  $\lambda \gg 1$  as obtained from (84) and (76), respectively, and from (67) which consists of the first three terms of (66)

for  $\lambda \rightarrow \infty$  and  $\lambda \rightarrow 0$ , respectively. Also shown in table 3 are tabulated values of the function

$$S(\lambda) = \lambda + \frac{1}{2} + \frac{1-\lambda}{2(1+\lambda)^3}, \tag{67}$$

which is obtained from (66) by retaining only the first term of the infinite series. Clearly, as pointed out by one of the referees, the expression given above provides a

very good approximation for  $S(\lambda)$  over the whole range of  $\lambda$ . We note that the values given by Keh & Chen (1993) for  $S(\frac{1}{2})$  and  $S(2)$  are 1.013 and 2.438, respectively, of which the former appears to be somewhat inaccurate.

It is worth remarking that the first two terms in (64) for  $S(\lambda)$  merely reflect the presence of the renormalization constraints. Specifically, since the motion of the bubbles in the suspension in the direction of  $\mathbf{H}$  must be accompanied by a corresponding mean back flux of fluid so that the constraint of zero mean flux at each point in the suspension is satisfied, a single bubble 1 in the fluid is then carried by the back flow with velocity  $-(c_1 U_1^{(0)} + c_2 U_2^{(0)})$ . This generates the leading term in (64). Similarly, the constraint on the mean temperature gradient at each point in the suspension requires that the average temperature gradient in the fluid be  $(1 - \frac{1}{2}c_1 - \frac{1}{2}c_2)\mathbf{H}$  since the temperature gradient inside an isolated bubble is  $\frac{3}{2}\mathbf{H}$  (Jeffrey 1973). In turn, this change in the average temperature gradient in the fluid is responsible for the second term in  $S(\lambda)$ . Clearly, in view of the results in table 3, the sum of the two renormalization constraints provides an excellent estimate for  $S(\lambda)$  when  $\lambda > \frac{1}{2}$ .

**5. Asymptotic expressions for  $S(\lambda)$  for the cases  $\lambda \gg 1$  and  $\lambda \ll 1$**

Although the series expression (66) can be used to evaluate  $S(\lambda)$  for all values of  $\lambda$ , its usefulness is limited since it converges very slowly when the two bubbles have drastically different sizes. In fact, when  $\lambda \rightarrow 0$  or  $\lambda \rightarrow \infty$  the thermal and hydrodynamic interactions are confined to a small region near the small bubble, and our method describing these interactions in terms of singularities at the centre of the large bubble is ineffective. So, it is desirable to find asymptotic expressions for  $S(\lambda)$  when  $\lambda \gg 1$  and  $\lambda \ll 1$ . A similar problem was studied by Chang & Acrivos (1986, 1987) for the case of heat conduction from a heated sphere to a matrix containing passive spheres of a different conductivity.

*5.1. Asymptotic expression for  $S(\lambda)$  when  $\lambda \gg 1$*

When  $\lambda \gg 1$ , the small test bubble 1 is immersed in a suspension of large bubbles 2. To find the average velocity of the test bubble  $\bar{U}_1$  from (63)–(65), we need to find the pair of mobility functions  $A_{11}$  and  $B_{11}$  which, according to (33) and (48), are the velocities of bubble 1 when the centreline is parallel and perpendicular to the temperature gradient, respectively, divided by  $U_1^{(0)}$  and when, in addition,  $U_2^{(0)} = 0$ , that is when bubble 2 is passive.†

When the two bubbles are far from each other, i.e. when  $R \gg \lambda$ , (45) and (53) yield the outer expansions

$$A_{11} = 1 - \frac{\lambda^3}{R^3} - 2 \frac{\lambda^3}{R^6} + O\left(\frac{\lambda^5}{R^8}\right), \quad l \gg O(\lambda), \quad \lambda \rightarrow \infty, \tag{68}$$

$$B_{11} = 1 + \frac{\lambda^3}{2R^3} + \frac{\lambda^3}{4R^6} + O\left(\frac{\lambda^5}{R^8}\right), \quad l \gg O(\lambda), \quad \lambda \rightarrow \infty, \tag{69}$$

where  $l \equiv R - \lambda$ .

Next, let us consider the case where the small bubble 1 is close to the large passive bubble 2, i.e. when  $l \sim O(1)$ . First, we estimate the order of magnitude of the velocity of the large passive bubble. According to Faxén’s law (Rallison 1978), it equals the

† A passive bubble is here defined as having its surface tension independent of temperature, i.e.  $d\gamma/dT = 0$ .

velocity at its centre induced by the singularities at the centre of the small active bubble. Since the latter is force-free, the induced velocity must decay at least as fast as  $O(a_1^2/r^2)$ , which contributes to  $A_{11}$  and  $B_{11}$  an  $O(1/\lambda^2)$  correction. But, since we wish to determine  $S(\lambda)$  only up to  $O(1/\lambda)$ , (63)–(65) show the the motion of the large bubble has only a smaller order effect on  $S(\lambda)$  so that the large bubble can be viewed as being stationary to this order of approximation.

Next, let us determine the inner expansions for  $A_{11}$ , i.e. the velocity of a small active bubble near a large passive one in the axisymmetric case, where the centreline is parallel to the temperature gradient. The temperature distribution in the absence of bubble 1 is (Acrivos *et al.* 1990):

$$T'_\infty = \left[ 1 + \frac{a_2^3}{2\rho_2^3} \right] \mathbf{H} \cdot \boldsymbol{\rho}_2.$$

Now, the presence of the small bubble 1 induces a temperature disturbance  $T'$  such that  $\partial(T' + T'_\infty)/\partial\rho_1 = 0$  is satisfied on the surface of the small bubble. Expanding  $T'_\infty$  about the centre of the small bubble, this boundary condition becomes

$$\frac{\partial T'}{\partial\rho_1} = -\frac{\partial}{\partial\rho_1} \left[ \frac{3l}{\lambda} \mathbf{H} \cdot \boldsymbol{\rho}_1 - \frac{3}{4a_1\lambda} \frac{\mathbf{H} \cdot \mathbf{r}}{r} \left( 3 \frac{\mathbf{H}\mathbf{H}}{H^2} - \mathbf{I} \right) : \boldsymbol{\rho}_1 \boldsymbol{\rho}_1 + O\left(\frac{1}{\lambda^2}\right) \right] \quad \text{at } \rho_1 = a_1, \quad (70)$$

while the boundary condition on the surface of the large bubble is still  $\partial T'/\partial\rho_2 = 0$  at  $\rho_2 = a_2$  since  $\partial T'_\infty/\partial\rho_2 = 0$ .

Next we examine the effect of each term in the above expansion separately, noting that, within the inner region  $1 \leq l < O(\lambda)$ , the large bubble can be replaced, as far as its first-order effects on the small bubble are concerned, with a non-conducting, stress-free planar wall.

The first term in the bracket of the right-hand side of (70) corresponds to the case when a non-conducting bubble has been placed near a plane on which the temperature gradient perpendicular to it has been set equal to  $3lH/\lambda$ . But as shown in §2, the velocity of the bubble is then the same as that of an isolated bubble in a linearly varying ambient temperature field, hence

$$\mathbf{U}_1 = \frac{3l}{\lambda} \mathbf{U}_1^{(0)}.$$

On the other hand, the velocity corresponding to the second term on the right-hand side of (70) can be obtained using the procedure of §3. Specifically, by the method of images, the plane can be replaced by an image bubble so that both the temperature and the velocity fields are symmetric about the plane thereby satisfying identically the non-conducting and stress-free boundary conditions on the plane. The problem then becomes that of determining the motion of two identical non-conducting bubbles in an ambient temperature field  $T'_\infty$ , which is

$$T'_\infty = -\frac{3}{4a_1\lambda} \frac{\mathbf{H} \cdot \mathbf{r}}{r} \left( 3 \frac{\mathbf{H}\mathbf{H}}{H^2} - \mathbf{I} \right) : \boldsymbol{\rho}_1 \boldsymbol{\rho}_1 \quad \text{at } \rho_1 = a_1,$$

on the side of the original bubble and symmetrically on the side of the image bubble. In turn, the velocities of the two bubbles are calculated by applying the technique of §3 to the solution of the above problem, which gives for the velocity of bubble 1

$$\mathbf{U}_1 = \mathbf{U}_1^{(0)} \left\{ \frac{1}{\lambda} \sum_{n=2}^{\infty} \frac{C_n}{l^n} \right\}, \quad (71)$$

where the  $C_n$ , with  $C_2 = -\frac{3}{4}$ ,  $C_3 = C_4 = 0$ ,  $C_5 = -\frac{3}{16}$ ,  $C_6 = 0$ ,  $C_7 = -\frac{9}{64}$ , ..., are constants found by solving the two-bubble problem.

Finally, combining the contributions from the two terms, we obtain

$$A_{11} = \frac{3l}{\lambda} + \frac{1}{\lambda} \sum_{n=2}^{\infty} \frac{C_n}{l^n} + O\left(\frac{1}{\lambda^2}\right), \quad 1 \leq l < O(\lambda), \quad \lambda \rightarrow \infty. \quad (72)$$

We next apply the same method to determine the inner expansion for  $B_{11}$ , i.e. the velocity of a small active bubble near a large passive one when  $\mathbf{H}$  is perpendicular to the centreline of the two bubbles.

Expanding  $T'_\infty$  about the centre of bubble 1, the boundary condition on the small bubble can be written as

$$\frac{\partial T'}{\partial \rho_1} = -\frac{\partial}{\partial \rho_1} \left[ \left( \frac{3}{2} - \frac{3l}{2\lambda} \right) \mathbf{H} \cdot \boldsymbol{\rho}_1 + \frac{3}{4a_1 \lambda} \left( \frac{\mathbf{H}r + r\mathbf{H}}{Hr} \right) : \boldsymbol{\rho}_1 \boldsymbol{\rho}_1 + O\left(\frac{1}{\lambda^2}\right) \right] \quad \text{at } \rho_1 = a_1. \quad (73)$$

The first term corresponds to the case where a bubble is placed near a non-conducting, stress-free wall with a constant temperature gradient parallel to the plane in the direction of  $\mathbf{H}$ . Once again, in view of the result of §2, the velocity of the bubble remains unchanged by the presence of the plane, i.e.

$$U_1 = \left[ \frac{3}{2} - \frac{3l}{2\lambda} \right] U_1^{(0)}.$$

The velocity corresponding to the second term can be found by a similar method as that used in the axisymmetric case to find (71). The result is

$$U_1 = U_1^{(0)} \left\{ \frac{1}{\lambda} \sum_{n=7}^{\infty} \frac{C'_n}{l^n} \right\}, \quad (74)$$

with  $C'_7 = \frac{9}{1024}, \dots$

Finally, we obtain

$$B_{11} = \left( \frac{3}{2} - \frac{3l}{2\lambda} \right) + \frac{1}{\lambda} \sum_{n=7}^{\infty} \frac{C'_n}{l^n} + O\left(\frac{1}{\lambda^2}\right), \quad 1 \leq l < O(\lambda), \quad \lambda \rightarrow \infty. \quad (75)$$

We now substitute the inner and outer asymptotic expansions for  $A_{11}$  and  $B_{11}$  into (65) to determine the asymptotic expression for  $S(\lambda)$ . To this end, we divide the interval of integration into an inner region  $\lambda + 1 \leq R \leq \lambda + O(\lambda)$  and an outer region  $\lambda + O(\lambda) \leq R \leq \infty$ , and note that within each domain we have a locally valid asymptotic expression for  $A_{11}$  and  $B_{11}$ .

By substituting (68) and (69) into (65) we find that, in the outer region,  $I_{out} = O(1/\lambda^2)$ , i.e. the contribution of the outer region to  $S(\lambda)$  is negligible to this order of approximation.

On the other hand, in the inner region, substituting (72) and (75) into (65), and noting that  $R$  is basically equal to  $\lambda$  in this region, we obtain

$$I_{in} = \frac{1}{\lambda} \sum_{n=2}^{\infty} \frac{C_n + C'_n}{n-1} + O\left(\frac{\ln \lambda}{\lambda^2}\right) = -\frac{0.84}{\lambda} + O\left(\frac{\ln \lambda}{\lambda^2}\right).$$

Here, since the sum containing the first  $k$  terms was found to be essentially linear in  $1/k$  for  $k > 20$ , the summation was determined by evaluating its first 30 terms and extrapolating the result.

Finally, by combining the results for the two regions, we obtain the following asymptotic result:

$$S(\lambda) = \lambda + \frac{1}{2} - \frac{0.84}{\lambda} + O\left(\frac{\ln \lambda}{\lambda^2}\right), \quad \lambda \rightarrow \infty. \quad (76)$$

Clearly, in view of (63), a bubble of type 1 could move *against* the temperature gradient for sufficiently large values of  $\lambda$  even if  $c_2$  is small.

### 5.2. Asymptotic expression for $S(\lambda)$ when $\lambda \ll 1$

We finally consider the other limiting case, i.e.  $\lambda = a_2/a_1 \ll 1$ , and find the average velocity of a large test bubble 1 immersed in a suspension of small bubbles 2. It turns out, however, that it is easier to determine the velocity of a large passive bubble in the presence of a small active one, i.e.  $A_{12}$  and  $B_{12}$  for the parallel and perpendicular cases, respectively.

When the two bubbles are far from each other, i.e. when  $R \gg 1$ , (45) and (53) yield the outer expansions:

$$A_{12} = \frac{\lambda^3}{R^3} + 2\frac{\lambda^2}{R^6} + O(\lambda^6), \quad R-1 \gg O(1), \quad \lambda \rightarrow 0, \quad (77)$$

$$B_{12} = -\frac{\lambda^3}{2R^3} - \frac{\lambda^3}{4R^6} + O(\lambda^6), \quad R-1 \gg O(1), \quad \lambda \rightarrow 0. \quad (78)$$

Next we derive the inner expansions for  $A_{12}$  and  $B_{12}$  when the small bubble 2 is close to the large bubble 1, i.e. when  $\bar{l} \sim O(1)$ , where  $\bar{l} \equiv (R-1)/\lambda$ . To begin with, we note that the velocity of a large passive bubble immersed in the flow field generated by a small active one can be determined using Faxén's law, in terms of the strength of the singularities at the centre  $O_2$  of the small bubble by evaluating the velocity they induce at the centre of the large bubble.

But, once again, since, up to leading order, the strengths of the singularities at  $O_2$  are affected only if  $\bar{l} \sim O(1)$ , these can be determined by replacing the passive bubble by a non-conducting, stress-free planar wall.

We start by considering the axisymmetric case. The strengths of the singularities at  $O_2$  can be evaluated by expanding the boundary conditions of the temperature field on the surface of the small bubble,

$$\frac{\partial T'}{\partial \rho_2} = -\frac{\partial}{\partial \rho_2} \left[ 3\lambda \bar{l} \mathbf{H} \cdot \boldsymbol{\rho}_1 - \frac{3\lambda}{4a_2} \frac{\mathbf{H} \cdot \mathbf{r}}{r} \left( 3 \frac{\mathbf{H}\mathbf{H}}{H^2} - \mathbf{I} \right) : \boldsymbol{\rho}_2 \boldsymbol{\rho}_2 + O(\lambda^4) \right] \quad \text{at} \quad \rho_2 = a_2. \quad (79)$$

The first term in the expansion corresponds to the constant-gradient case. Applying the method of images as in §5.1 we see that, in this case, the flow field is irrotational and decays as  $O(\lambda a_2^3/\rho_2^3)$ , thereby contributing to the velocity of bubble 1, or to  $A_{12}$  an  $O(\lambda^4)$ -term and to  $S(\lambda)$  an  $O(\lambda^2)$ -term, which is negligible to this order of approximation.

The second term in the temperature expansion is a quadratic distribution. The leading term of the flow field induced by bubble 2 is an  $O(\lambda a_2^2/\rho_2^2)$  stresslet term (Anderson 1985), which contributes to  $A_{12}$  an  $O(\lambda^4)$ -term and to  $S(\lambda)$  an  $O(\lambda)$ -term, while the contributions from higher-order singularities at the centre of bubble 2 (cf. (20)) are of smaller order and thus negligible. The strength of the stresslet at the centre of the small bubble or  $p_{01}^{(2)}$  can be found as an intermediate result in deriving (71). Finally, substitution of  $p_{01}^{(2)}$  into (20), yields  $U_1$  and then  $A_{12}$ ,

$$A_{12} = 3\lambda^3 + \lambda^3 \sum_{n=3}^{\infty} \frac{D_n}{l^n} + O(\lambda^4), \quad \bar{l} \sim O(1), \quad \lambda \rightarrow 0, \quad (80)$$

where the  $D_n$  are constants that can be found using the intermediate result in §5.1 with the first few of them being  $D_3 = \frac{3}{4}$ ,  $D_4 = D_5 = 0$ ,  $D_6 = \frac{3}{16}$ ,  $D_7 = 0$ ,  $D_8 = \frac{27}{256}$ , ...



Now let us turn to the perpendicular case, where the temperature expansion is

$$\frac{\partial T'}{\partial \rho_2} = -\frac{\partial}{\partial \rho_2} \left[ \left( \frac{3}{2} - \frac{3\bar{\lambda}}{2} \right) \mathbf{H} \cdot \boldsymbol{\rho}_2 + \frac{3\lambda}{4a_2} \left( \frac{\mathbf{H}\mathbf{r} + \mathbf{r}\mathbf{H}}{Hr} \right) : \boldsymbol{\rho}_2 \boldsymbol{\rho}_2 + O(\lambda^2) \right]. \quad (81)$$

Once again, the case corresponding to the constant-gradient term can be treated by the method of images and the flow field is found to be irrotational. This decays as  $O(a_2^3/\rho_2^3)$  which corresponds to a force quadrupole term and contributes to  $S(\lambda)$  an  $O(\lambda)$ -term, while the contributions of the higher-order singularities are of smaller order and thus negligible. In turn, the strength of this force quadrupole at the centre of the small bubble can be found using the intermediate results of §3.3 for identical bubbles.† Thus, on applying Faxén's law, the velocity of the large bubble is found to be

$$\mathbf{U}_1 = \mathbf{U}_2^{(0)} \left\{ -\frac{3}{4}\lambda^3 + \lambda^3 \sum_{n=3}^{\infty} \frac{E_n}{\bar{l}^n} + O(\lambda^4) \right\}, \quad \bar{l} \sim O(1), \quad \lambda \rightarrow 0, \quad (82)$$

with  $E_3 = -\frac{3}{8}$ ,  $E_4 = E_5 = 0$ ,  $E_6 = -\frac{3}{16}$ ,  $E_7 = 0$ ,  $E_8 = -\frac{3}{4}$ , ...

The second term in the temperature expansion (81) corresponds to a quadratic temperature distribution. The leading term of the velocity field induced by this singularity at the centre of the small bubble is an  $O(\lambda a_2^2/\rho_2^2)$  stresslet term, while all the other terms are of smaller order and can be neglected. However, this term does not contribute to  $\mathbf{U}_1$  which here is perpendicular to  $\mathbf{r}$ , since a stresslet only induces a velocity in the radial direction from its position. Therefore, we conclude that the effect of the second term on  $\mathbf{U}_1$  is smaller than  $O(\lambda^3)$ , so that its contribution to  $S(\lambda)$  is negligible. Therefore,

$$\mathbf{B}_{12} = -\frac{3}{4}\lambda^3 + \lambda^3 \sum_{n=3}^{\infty} \frac{E_n}{\bar{l}^n} + O(\lambda^4), \quad \bar{l} \sim O(1), \quad \lambda \rightarrow 0. \quad (83)$$

Now, as in §5.1 we use the inner and outer expansions for  $A_{12}$  and  $B_{12}$ , (77), (78) (80) and (83) to evaluate the integral in (65) and obtain the asymptotic expression for  $S(\lambda)$ . First, we divide the interval of integration into an inner region  $1 + \lambda \leq R \leq 1 + R_1$  and an outer region  $1 + R_1 \leq R \leq \infty$  with  $R_1 \sim O(1)$ , but note that the integral in each region will depend on the value of  $R_1$ . This difficulty can be circumvented by constructing a uniformly valid expression for the integrand (Van Dyke 1964) or equivalently rewriting (65) as

$$\int_{1+\lambda}^{\infty} f_{out}(R) dR + \int_{1+\lambda}^{R_1} [f_{in}(R) - f_{out}(R)] dR, \quad \lambda \rightarrow 0,$$

and letting  $R_1 \rightarrow \infty$ , where  $f_{in}(R)$  and  $f_{out}(R)$  refer to the integrand in (65) evaluated using, respectively, the inner and outer expansions for  $A_{12}$  and  $B_{12}$ . Upon evaluating the sum by the same method as that used in arriving at (76), we find that

$$S(\lambda) = 1 - 0.57\lambda + O(\lambda^2), \quad \lambda \rightarrow 0. \quad (84)$$

This result for  $S(\lambda)$  can be interpreted in terms of the effective continuum approach (Acrivos & Chang 1986) by noting that the suspension of small bubbles of vanishing size acts as an effective continuum with effective viscosity  $\mu^* = \mu(1 + c_2)$  and effective conductivity  $k^* = k(1 - \frac{3}{2}c_2)$ , with  $k$  being the conductivity of the pure fluid. But a large bubble immersed in such an effective continuum will move, according to (9), with

† In this case, although from (13) the velocities of the bubbles are the same as if they were isolated, the strength of the force quadrupole depends on the distance between the two bubbles.

velocity  $(1 - c_2) U_1^{(0)}$ , in agreement with the first term in (84), which therefore reflects the effect of the increase in the viscosity of the surrounding fluid, due to the presence of the small bubbles, on the velocity of the large bubble.

As noted earlier, the function  $S(\lambda)$  is seen plotted in figure 1 together with its two asymptotic expressions as obtained for (84) and (76) for, respectively,  $\lambda \rightarrow 0$  and  $\lambda \rightarrow \infty$ .

This work was supported in part by the National Science Foundation grant CTS-9012937.

#### REFERENCES

- ACRIVOS, A. & CHANG, E. 1986 The transport properties of non-dilute suspensions. Renormalization via an effective continuum method. In *Physics and Chemistry of Porous Media II* (ed. J. R. Banavar, J. Koplik & K. W. Winkler), pp. 129–142. AIP Conf. Proc. 154. New York: AIP.
- ACRIVOS, A., JEFFREY, D. J. & SAVILLE, D. A. 1990 Particle migration in suspensions by thermocapillary or electrophoretic motion. *J. Fluid Mech.* **212**, 95–110.
- ANDERSON, J. L. 1985 Droplet interactions in thermocapillary motion. *Intl J. Multiphase Flow* **11**, 813–824.
- CHANG, E. & ACRIVOS, A. 1986 Rate of heat conduction from a heated sphere to a matrix containing passive spheres of a different conductivity. *J. Appl. Phys.* **59**, 3375–3382.
- CHANG, E. & ACRIVOS, A. 1987 Conduction of heat from a planar wall with uniform surface temperature to a monodispersed suspension of spheres. *J. Appl. Phys.* **62**, 771–776.
- HAPPEL, J. & BRENNER, H. 1965 *Low Reynolds Number Hydrodynamics*. Prentice-Hall.
- JEFFREY, D. J. 1973 Conduction through a random suspension of spheres. *Proc. R. Soc. Lond. A* **335**, 355–367.
- JEFFREY, D. J. & ONISHI, Y. 1984 Calculation of the resistance and mobility functions for two unequal rigid spheres in low-Reynolds-number flow. *J. Fluid Mech.* **139**, 261–290.
- KEH, H. J. & CHEN, S. H. 1990 The axisymmetric thermocapillary motion of two fluid droplets. *Intl J. Multiphase Flow* **16**, 515–527.
- KEH, H. J. & CHEN, L. S. 1992 Droplet interactions in axisymmetric thermocapillary motion. *J. Colloid Interface Sci.* **151**, 1–16.
- KEH, H. J. & CHEN, L. S. 1993 Droplet interactions in thermocapillary migration. *Chem. Engng Sci.* (to be published).
- MEYYAPPAN, M. & SUBRAMANIAN, R. S. 1984 The thermocapillary motion of two bubbles oriented arbitrarily relative to a thermal gradient. *J. Colloid Interface Sci.* **97**, 291–294.
- MEYYAPPAN, M., WILCOX, W. R. & SUBRAMANIAN, R. S. 1983 The slow axisymmetric motion of two bubbles in a thermal gradient. *J. Colloid Interface Sci.* **94**, 243–257.
- RALLISON, J. M. 1978 Note on the Faxén relations for a particle in Stokes flow. *J. Fluid Mech.* **88**, 529–533.
- SATRAPE, J. V. 1992 Interactions and collisions of bubbles in the thermocapillary motion. *Phys. Fluids A* **4**, 1883–1900.
- SUBRAMANIAN, R. S. 1985 The Stokes force on a droplet in an unbounded fluid medium due to capillary effects. *J. Fluid Mech.* **153**, 389–400.
- VAN DYKE, M. 1964 *Perturbation Methods in Fluid Mechanics*. Academic.
- THOVERT, J. F. & ACRIVOS, A. 1989 The effective thermal conductivity of a random polydisperse suspension of spheres to order  $C^2$ . *Chem. Engng Commun.* **82**, 177–191.
- YOUNG, N. O., GOLDSTEIN, J. S. & BLOCK, M. J. 1959 The motion of bubbles in a vertical temperature gradient. *J. Fluid Mech.* **6**, 350–356.