J. Fluid Mech. (2001), *vol.* 443, *pp.* 377–401. Printed in the United Kingdom

Thermocapillary migration of bubbles: convective effects at low Péclet number

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(Received 23 May 2000 and in revised form 17 April 2001)

The effect of a weak convective heat transfer on the thermocapillary interaction of two bubbles migrating in an externally imposed temperature gradient is examined. It is shown that, for short and moderate separation distances, the corrections to the individual migration velocities of the bubbles are of $O(Pe)$, where Pe is Péclet number. For separation distances larger than $O(Pe^{-1/2})$ the correction is of $O(Pe^2)$ as previously found for an isolated drop. The perturbations to the bubble velocities have opposite signs: the motion of the leading bubble is enhanced while the motion of the trailing one is retarded. A newly found feature is that equal-sized bubbles, which otherwise would move with equal velocities, acquire a relative motion apart from each other under the influence of convection. For slightly unequal bubbles there are three different regimes of large-time asymptotic behaviour: attraction up to the collision, infinite growth of the separation distance, and a steady migration with equal velocities, the steady motion separation distance being a function of the parameters of the problem. Sufficient conditions for the realization of each regime are given in terms of the Péclet number, initial separation and radii ratio.

1. Introduction

The hydrodynamic interaction between drops, bubbles and particles is very important in various multiphase flows. Analyses of this interaction acquire qualitatively new features caused by the phenomenon of thermocapillarity, i.e. by the dependence of the interfacial tension on temperature. Thermal gradients that appear in the system in the presence of heat transfer induce surface tension gradients that, in turn, generate thermocapillary flow in the surrounding fluid. This flow can drastically change the motion of fluid particles in multiphase systems. Thermocapillary-induced migration of drops and bubbles under an externally imposed thermal gradient in the surrounding fluid was first described by Young, Goldstein & Block (1959). It has since attracted wide attention and, in particular in recent years, it has become relevant in applications to materials processing under conditions of micro-gravity. A review of the subject is given by Subramanian (1992) and by Wozniak, Siekmann & Srulijes (1988).

Most of the theoretical studies of the effect were conducted under the assumption of negligible inertia and convective transport, i.e. negligibly small Reynolds, *Re*, and Péclet, Pe, numbers. The Péclet number in thermocapillary motion is often addressed through the Marangoni number. Within this approximation of Stokes flow and a harmonic temperature field, the problem of the thermocapillary migration of two spherical drops is similar to classical problems describing their hydrodynamic interaction (Stimson & Jeffery 1926; Happel & Brenner 1965; Haber, Hetsroni &

Solan 1973). In this limit the motion is quasi-stationary and the velocities of two drops are found as functions of the instantaneous geometric configuration, drop radius ratio and the material properties of the fluids. The axisymmetric case of two bubbles moving along their line of centres in a thermal gradient was treated by Meyyappan, Wilcox & Subramanian (1983), who found that equal-sized bubbles exert no influence on each other's velocity for all separation distances. These findings were later confirmed analytically by Feuillebois (1989). Anderson (1985) obtained the same result in the more general case of a thermocapillary migration of two arbitrarily oriented bubbles with respect to an applied temperature gradient using the approximate method of reflections, while Acrivos, Jeffery & Saville (1990) showed that this result prevails when considering a monodisperse collection of interacting bubbles. This exact cancellation of the interaction effects is likely to be a consequence of a high symmetry of the linear problem, where the disturbances due to the thermal and hydrodynamic fields neutralize each other.

In the nonlinear limit, where the effect of convective transport or inertia becomes non-negligible, analytical results were reported mainly for the case of thermocapillary migration of an isolated particle. The influence of a weak convective transport, i.e. small but non-zero Péclet number, on the thermocapillary migration of a single particle was studied by Subramanian (1981) for the case of a bubble, and by Subramanian (1983) for the case of a drop. The effects of inertia and interface deformations were neglected, i.e. the Reynolds and capillary, Ca, numbers were presumed to be zero. Bratukhin (1975, 1977) examined these effects, assuming that they are of the same order of magnitude, with Pe, Re and Ca being proportional to one controlling parameter, the Marangoni number. The singular perturbations analysis showed that the corrections to the temperature and velocity fields are of $O(Pe)$, but the correction to the migration velocity is of $O(Pe^2)$. Balasubramaniam & Subramanian (1996) considered the steady-state thermocapillary migration of a single bubble at large Pe and two limiting cases of $Re = 0$ and $Re \to \infty$ and showed that for both limiting cases the migration velocity approaches a constant value as $Pe \to \infty$.

In experiments there is always a non-negligible convective transport and it is particularly interesting to know if this remarkable effect of cancellation, mentioned above for equal-sized bubbles, extends to a weakly nonlinear limit of small Pe. It is predicted that this effect does not hold and the convective transport around the interacting bubbles may lead to non-symmetric contributions to their individual velocities resulting in relative motion.

The literature on the convective transport effect for interacting drops and bubbles is limited. We have recently analysed the problem of the spontaneous migration of two drops induced by interphase surfactant (heat) transfer, in the absence of an externally imposed gradient, for negligible Re and finite, but small Pe (Lavrenteva, Leshansky & Nir 1999). It was shown that the correction to the migration velocity of a drop at moderate initial separation distances is of order $\sqrt{Pe}$. Balasubramaniam & Subramanian (1999) have studied the migration of two bubbles in a uniform temperature gradient and have shown that, in this case, the interaction of the trailing bubble with the thermal wake of the leading one substantially retards its migration speed. No analytical studies of the convective transport effect on the thermocapillary migration of interacting particles in an external gradient field at small Pe and Re have been reported so far.

In the present paper we extended the analysis of Subramanian (1981) to the case of two bubbles interacting in an external temperature gradient. For moderate separation distances the correction to the quasi-stationary velocities of the bubbles is found to

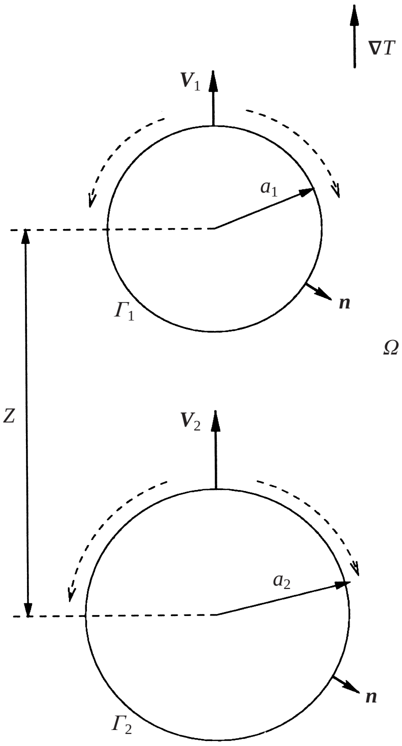

FIGURE 1. Two bubbles migrating in a temperature gradient.

be proportional to Pe. For the asymptotic case of large initial separation distance between the bubbles, $Z \gg Pe^{-1/2}$, the leading-order correction term is expected to be of order Pe^2 as in the case of a single bubble (Subramanian 1981). The results of the present paper show that the convective effect in the case of thermocapillary motion of two bubbles in an external temperature gradient is more pronounced than that in the case of single drop (bubble) migration and less pronounced than in the case of spontaneous drop interaction caused by interphase mass (heat) transport.

Our analysis predicts that a weak convective transport results in a relative motion of equal-sized bubbles apart from each other and qualitatively changes the interaction pattern in the case of unequal bubbles. When the leading bubble is smaller than the trailing one and the radii ratio is large enough, the separation distance decreases up to the collision of the bubbles as would happen were conduction is the only transport mechanism. In contrast to this, if the radii ratio is close enough to unity, the motion tends with time to a steady asymptotic state, when the bubbles translate with equal constant velocity, the separation distance being a function of Péclet number and radii ratio.

2. Statement of the problem

Consider two bubbles of radii a_1 and a_2 submerged in an unbounded viscous Newtonian fluid which is quiescent at infinity and has a constant uniform thermal gradient, $T = \boldsymbol{A} \cdot \boldsymbol{x}$, far away from the bubbles, as shown in figure 1. The thermal diffusivity, density and the viscosity of the liquid are χ_o, ρ_o and η_o, respectively. Gravity and buoyancy forces are absent. The viscosity, density and thermal conductivity of the gas phase are assumed negligible compared to the same properties in the liquid. Thus, only the governing equations for the liquid need to be considered. It is supposed that the magnitude of $\boldsymbol{A}$ is small and the changes in temperature do not affect any physical properties of the liquid in the bulk and at the interface, except for the interfacial tension which varies linearly with temperature: $\sigma = \sigma_0 - \sigma_1 T$, where σ_0 and σ_1 are positive constants.

The following scaling is chosen: the radius of the leading bubble, a_1, for length, $u = \eta_o^{-1}\sigma_1 A a_1$ for velocity, a_1/u for time, $\sigma_1 A$ for pressure and $A a_1$ for temperature. Let $\boldsymbol{x} = (x_1, x_2, x_3)$ be a dimensionless radius vector to a point in the laboratory coordinate system with some chosen origin. In this coordinate system let $\boldsymbol{A} = (0, 0, A)$. Thus, the dimensionless velocity field $\boldsymbol{v} = (v_1, v_2, v_3)$, the pressure p and the temperature field Θ are governed by the following equations:

$$Pe(\partial\Theta/\partial t + \boldsymbol{v}\cdot\nabla\Theta) = \Delta\Theta, \tag{1}$$

$$Re\,(\partial\boldsymbol{v}/\partial t + \boldsymbol{v}\cdot\nabla\boldsymbol{v}) = -\nabla p + \Delta\boldsymbol{v}, \tag{2}$$

$$\nabla\cdot\boldsymbol{v} = 0, \quad \boldsymbol{x}\in\Omega, \tag{3}$$

and the boundary conditions

$$\boldsymbol{\Pi}\cdot\boldsymbol{n} = (Ca^{-1} - \Theta)K\boldsymbol{n} - \nabla_\tau\Theta, \tag{4}$$

$$\boldsymbol{v}\cdot\boldsymbol{n} = V_{ni}, \tag{5}$$

$$\partial\Theta/\partial n = 0, \quad \boldsymbol{x}\in\Gamma_i, \tag{6}$$

$$\boldsymbol{v}\to\boldsymbol{0}, \quad \Theta\to x_3 \quad \text{as} \quad |\boldsymbol{x}|\to\infty, \tag{7}$$

where Ω denotes the domain occupied by the liquid, Γ_i is the surface of bubble i, V_{ni} is the normal interfacial velocity of bubble i, $\boldsymbol{n}$ is an outer normal unit vector to a corresponding bubble surface, $\boldsymbol{\Pi} = -p\boldsymbol{I} + (\nabla\boldsymbol{v} + (\nabla\boldsymbol{v})^T)$ is the stress tensor, K is a curvature and $\nabla_\tau = \nabla - \boldsymbol{n}(\boldsymbol{n}\cdot\nabla)$ denotes the surface gradient. The dimensionless parameters of this problem are: the Péclet number $Pe = ua_1/\chi_o$, the Reynolds number $Re = ua_1\rho_o/\eta_o$ and the capillary number $Ca = \sigma_1 A a_1/\sigma_0$. If Ca is negligible it follows from (4) that the bubbles preserve their spherical shape with radii $R_i = a_i/a_1$ and that $V_{ni} = \boldsymbol{V}_i\cdot\boldsymbol{n}$, where $\boldsymbol{V}_i$ is velocity of bubble i. The total force acting on each bubble is given by

$$\boldsymbol{F}_i = \oint_{\Gamma_i}\boldsymbol{\Pi}\cdot\boldsymbol{n}\,\mathrm{d}s = 0, \quad i = 1, 2. \tag{8}$$

The problem is completed by the kinematic condition

$$\boldsymbol{V}_i = \mathrm{d}\boldsymbol{Z}_i(t)/\mathrm{d}t, \quad i = 1, 2, \tag{9}$$

where $\boldsymbol{Z}_i(t)$ denotes a radius vector to the centre of bubble i at moment t, and by the initial conditions

$$\boldsymbol{Z}_i(0) = \boldsymbol{Z}_{i0}, \quad i = 1, 2, \tag{10}$$

$$\Theta\,(0, \boldsymbol{x}) = \Theta_0(\boldsymbol{x}), \quad \boldsymbol{x}\in\Omega. \tag{11}$$

If the bubbles are placed initially parallel to the x_3-axis, then, in view of the form of $\boldsymbol{A}$, the problem (2)–(11) possesses axial symmetry. Let the origin of the coordinate system be chosen on the line connecting the bubble centres, then $\boldsymbol{Z}_i = Z_i\boldsymbol{e}_3$ and $\boldsymbol{V}_i = V_i\boldsymbol{e}_3$. In this paper we consider only the axisymmetric case.

If $Re = 0$, $Ca = 0$ and $Pe = 0$, the problem (2)–(11) reduces to the classical quasi-steady formulation investigated by Meyyappan *et al.* (1983) and Feuillebois (1989). A brief summary of the methods used in these papers and their results is given in the next section. Our aim is to study the effect of a weak convective transport (with small but non-zero Pe), assuming non-deformability of the bubbles and negligible inertia.

3. Zero-order approximation

The zero-order approximation is the solution of (2)–(11) under the following assumptions: $Re = 0$, $Ca = 0$ and $Pe = 0$. In this case the thermal and hydrodynamic problems decouple. The temperature field can be found independently from the velocity field as a harmonic function in Ω satisfying the boundary conditions (6), (7). Once the temperature distribution on the bubble interfaces is known, the velocity field is determined by solving (2)–(5), (8). The positions of the bubbles, Z_1 and Z_2, and the separation distance between their centres, $Z(t) = Z_1(t) - Z_2(t)$, change with time. Although the temperature and velocity fields are found as solutions of non-evolutionary problems, they depend on time parametrically via the quasi-steady evolution of the geometry.

The quasi-stationary problem was first treated by Meyyappan *et al.* (1983) by use of bi-spherical coordinates. The solution of the thermal problem was obtained as a series involving coefficients to be calculated numerically from the infinite linear system of equations. A method that allows that numerical effort of Meyyappan *et al.* (1983) to be avoided and leads to a closed-form solution of the problem was used by Feuillebois (1989), Loewenberg & Davis (1995) and Barton & Subramanian (1990). This method is based on the introduction of a scalar heat potential and it was first proposed by Sadhal (1983). Furthermore, we adopt the technique of the heat potential to construct a zero-order approximation to our problem, and the technique of Meyyappan *et al.* (1983) to construct the first-order expansion term. The details of this procedure are given in Appendix A.

Let (r, θ, ϕ) be a spherical coordinate system with origin coinciding with the origin of a bi-spherical coordinate system at some moment t. The temperature and the stream function far from the bubbles have the following asymptotic forms:

$$\Theta^{(0)} - x_3 = a(Z, R)\mathscr{P}_1(\mu)/2r^2 + O(r^{-3}), \qquad (12)$$

$$\psi^{(0)} = -b(Z, R)\mathscr{C}_3^{-1/2}(\mu) + O(r^{-1}), \qquad (13)$$

where the size ratio $R = R_2 = a_2/a_1$, $\mu \equiv \cos\theta$, $\mathscr{P}_1(\mu)$ and $\mathscr{C}_3^{-1/2}(\mu)$ denote Legendre and Gegenbauer polynomials respectively, a and b are constants given by (A 14) and (A 13), which depend on the separation and the bubble radii. Note that it follows from (12) that the far-field distortion of temperature due to each bubble is a thermal dipole.

The constants a and b can be determined from the formal expansion of the quasi-steady solutions for the temperature and the stream function at infinity (see Appendix A) or from the large-Z asymptotic solution. The dependence of these constants on the separation distance for different bubble radii is shown in figure 2(*a*–*d*). In the case of equal-sized bubbles, when $R = 1$, it follows from the formulas of Appendix A and the symmetry of the problem that $b(Z) = 0$. Hence, the leading term of the velocity decays as $1/r^3$ at $r \to \infty$. It was shown by Feuillebois (1989) that, in this case, the bubbles move with constant equal speeds and, hence, the separation distance does not depend on time. For unequal bubbles, the calculations performed by Meyyappan *et al.* (1983) demonstrate that the larger bubble always migrate with larger velocity. The distance between the bubbles grows with time if

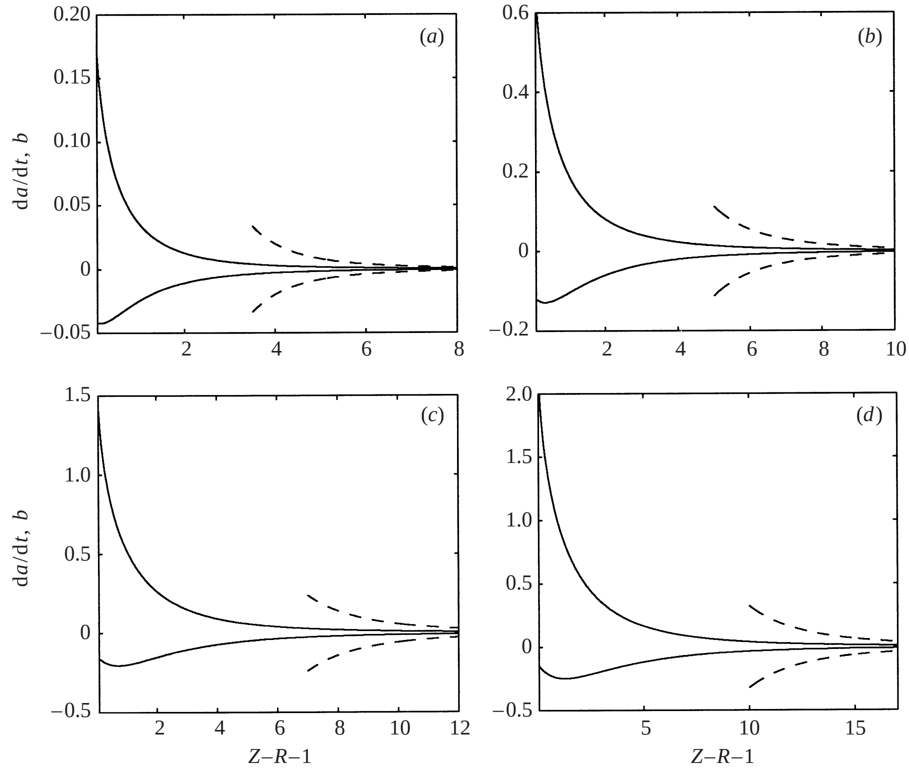

FIGURE 2. The dependence of $\dot{a}(Z(t))$ (lower curve) and $b(Z(t))$ (upper curve) on the separation distance, $Z - R - 1$, for different radii ratio: (*a*) $R = 1.5$, (*b*) $R = 2.5$, (*c*) $R = 4$, (*d*) $R = 6$. Dashed curves are asymptotic evaluations for large Z (equation (32)).

the larger bubble is a leading, $R < 1$, and it decreases in the opposite case, $R > 1$. Thus, as far as the relative motion of bubbles is concerned, $R = 1$ can be considered as a critical value, in which the interaction pattern qualitatively changes. It can be expected that in the vicinity of this critical value even small perturbations of the other parameters substantially change the motion pattern. Below we demonstrate that this is actually the case when convective transport is not entirely negligible and when small perturbations of the Péclet number are considered.

4. Construction of the solution for small Pe

In this section we construct the first correction term to the quasi-stationary solution described above when $0 < Pe \ll 1$. Specifically, we are interested in the first correction to the temperature field $\Theta = \Theta^{(0)} + \epsilon(Pe)\Theta^{(1)} + o(\epsilon(Pe))$ and to the bubble migration velocities, $V_i = V_i^{(0)}(Z) + \delta(Pe)V_i^{(1)} + o(\delta(Pe))$, $i = 1, 2$, where $\delta(Pe)$, $\epsilon(Pe) \ll 1$. It is easy to see that an attempt to use a regular perturbation technique with the natural choice, $\epsilon = Pe$, leads to a problem for the leading-order correction to the temperature $\Theta^{(1)}$ which does not possess solutions that vanish at infinity. Thus, one has to apply singular perturbation methods. Following a well-established procedure (Acrivos & Taylor 1962) we construct inner and outer expansions for the temperature field disturbance, $\Theta^* = \Theta - x_3$, denoted by h and H, respectively; $h(t, \boldsymbol{x})$, $t > 0$, $\boldsymbol{x} \in \Omega$ satisfies the boundary condition $\partial h/\partial n = -\partial x_3/\partial n$, $\boldsymbol{x} \in \partial\Omega$ and $H(t, \eta)$, $t > 0$,

$\boldsymbol{\eta} \in \mathscr{R}^3 \backslash \{\mathbf{0}\}$, $\boldsymbol{\eta} = \varepsilon \boldsymbol{x}$, vanishes at infinity

$$\lim_{|\boldsymbol{\eta}|\to\infty} H = 0, \tag{14}$$

while the two expansions match asymptotically

$$H(|\boldsymbol{\eta}| \to 0) = h(|\boldsymbol{x}| \to \infty). \tag{15}$$

4.1. *Outer expansion*

Let $\rho = \varepsilon r = |\boldsymbol{\eta}|$ and let $\Theta^*(t, r, \mu) = H(t, \rho, \mu)$, then

$$\Delta_\rho H = \varepsilon^{-2} Pe(H_t + \varepsilon \boldsymbol{U} \cdot \nabla_\rho H + U_3), \tag{16}$$

with $\boldsymbol{U}(\boldsymbol{\eta}) = \boldsymbol{v}(\boldsymbol{\eta}/\varepsilon)$ and $U_3(\boldsymbol{\eta}) = v_3(\boldsymbol{\eta}/\varepsilon)$. Recall that the velocity field satisfies the steady Stokes equations with no external body force and hence it decays as $1/r^2$ at $r \to \infty$. It is readily seen from (16) that at a length scale $O(Pe^{-1/2})$ the conduction and convection terms balance each other. Thus, an appropriate choice for the outer variable is $\varepsilon = Pe^{1/2}$. It means that in the outer region $r \geqslant O(Pe^{-1/2})$. The quasi-stationary solution described in the previous section can only be used as a zero-order expansion term if both bubbles lie in the inner region, i.e. the separation distance $Z \leqslant O(Pe^{-1/2})$. Otherwise, the interaction of each bubble with an external thermocapillary flow will dominate over their mutual interaction, and an $O(Pe^2)$ correction predicted by Subramanian (1981) is expected to be the leading order. We expand H as $H = f_0(\varepsilon)H^{(0)} + f_1(\varepsilon)H^{(1)} + o(f_1(\varepsilon))$ and substitute in (16). It follows that the zero-order outer expansion term $H^{(0)}(t, \rho, \mu)$ satisfies the non-homogeneous heat equation

$$f_0(\varepsilon)\Delta_\rho H^{(0)} = f_0(\varepsilon)H_t^{(0)} + U_3^{(0)}(t, \rho, \mu), \quad U_3^{(0)} = \varepsilon^2 b(t)\mu P_2(\mu)/\rho^2 + O(\varepsilon^3/\rho^3), \tag{17}$$

with the following boundary conditions:

$$\lim_{\rho\to\infty} H^{(0)} = 0, \quad \lim_{\rho\to 0} f_0(\varepsilon)\rho^2 H^{(0)}/\varepsilon^2 = \lim_{r\to\infty} r^2 h^{(0)} = a(t)\mu/2. \tag{18}$$

Consequently, we find from (17), (18) that $f_0(\varepsilon) = \varepsilon^2$.

The solution of the formulated problem can be expressed as a superposition $H^{(0)} = H_1^{(0)} + H_2^{(0)}$, where $H_1^{(0)}$ satisfies the homogeneous heat equation with boundary conditions (18) and $H_2^{(0)}$ satisfies equation (17) with corresponding homogeneous boundary conditions. $H_1^{(0)}$ has the form

$$H_1^{(0)}(t, \rho, \mu) = \frac{\rho\mu}{8\sqrt{\pi}} \int_0^t \frac{a(\tau)}{(t-\tau)^{5/2}} \exp\left\{-\frac{\rho^2}{4(t-\tau)}\right\} \mathrm{d}\tau, \tag{19}$$

where $a(t) = \dot{a}(t) = 0$ at $t \to 0$, and where the thin temporal boundary layer near $t = 0$ is neglected.

Let $\hat{b}(s)$ be the Laplace image of $b(t)$, then the solution of the second problem is the inverse Laplace image of

$$\hat{H}_2^{(0)} = \hat{b}(s)\left[\frac{2(-1 + \mathrm{e}^{-\rho\sqrt{s}}(1 + \rho\sqrt{s}))\mu}{5\rho^2 s}\right.$$
$$\left. - \frac{\mathrm{e}^{-\rho\sqrt{s}}(30 - 30\mathrm{e}^{\rho\sqrt{s}} + 30\rho\sqrt{s} + 12\rho^2 s + 3\mathrm{e}^{\rho\sqrt{s}}\rho^2 s + 2\rho^3 s^{3/2})P_3(\mu)}{5\rho^4 s^2}\right]. \tag{20}$$

Expanding the zero-order outer solution up to $O(\varepsilon^2)$ near $\rho = 0$ and re-writing it in terms of the inner variable we obtain

$$H = \frac{\mu a(t)}{2r^2} - \varepsilon^2 \left(\frac{\mu \dot{a}(t)}{4} + \frac{\mu(1+\mu^2)b(t)}{8} \right) + O(\varepsilon^3). \tag{21}$$

The form of (21) indicates that the second term of the inner expansion is of the order of ε^2:

$$h = h^{(0)} + \varepsilon^2 h^{(1)} + \cdots . \tag{22}$$

In the case of equal-sized bubbles $b(t) = 0$ and a does not depend on time, hence the expansion of the outer solution reads

$$H = \frac{\mu a}{2r^2} + O(\varepsilon^3),$$

whereas the inner expansion has the form (22) with a non-zero $h^{(1)}$, as a result of the convective transport in the vicinity of the bubbles.

4.2. *Inner expansion*

The first term of the inner expansion is given by the quasi-stationary temperature, $h^{(0)} = \Theta^{(0)} - x_3$. The second term, $h^{(1)}$, can be found as a solution of the following boundary value problem:

$$\Delta_r h^{(1)} = \Xi\,(\boldsymbol{x}, t), \quad \boldsymbol{x} \in \Omega, \tag{23}$$

$$\partial h^{(1)}/\partial n = 0, \quad \boldsymbol{x} \in \partial\Omega, \tag{24}$$

$$\lim_{|\boldsymbol{x}|\to\infty} h^{(1)} = -\left(\frac{\mu \dot{a}(t)}{4} + \frac{\mu(1+\mu^2)b(t)}{8} \right), \tag{25}$$

where

$$\Xi\,(\boldsymbol{x}, t) = h_t^{(0)} + \boldsymbol{v}^{(0)} \cdot \nabla_r h^{(0)} + v_3^{(0)}.$$

The term proportional to $\dot{a}(t)$ and to $b(t)$ in (25) reflects the influence of the temporal change in the mutual positions of the bubbles, and the advection from the outer region, respectively, and the right-hand side of (23) corresponds to the convective transport in their vicinity.

Since the zero-order solution $h^{(0)}, \boldsymbol{v}^{(0)}$ is found in the bi-spherical coordinate system (ξ, ζ) fixed with the travelling bubbles (see Appendix A), it is a natural choice to solve (23)–(25) in this coordinate system. The interface of the first bubble is described by the coordinate surface $\xi = \alpha > 0$ and the interface of the second bubble corresponds to $\xi = -\beta < 0$. In this case $h^{(0)}(\boldsymbol{x}, t) = h^{(0)}(\xi(\boldsymbol{x}, t), \zeta(\boldsymbol{x}, t), Z^{(0)}(t))$, so that care needs to be exercised when computing the term $h_t^{(0)}$ on the right-hand side of (23). Finally, this reduces to

$$h_t^{(0)} = -\frac{V^*}{c}\left((1 - \tilde{\mu}\cosh\xi)\frac{\partial h^{(0)}}{\partial \xi} + \sinh\xi(1-\tilde{\mu}^2)\frac{\partial h^{(0)}}{\partial \tilde{\mu}} \right)$$
$$+ \frac{\dot{c}}{c}\left(\tilde{\mu}\sinh\xi\frac{\partial h^{(0)}}{\partial \xi} - \cosh\xi(1-\tilde{\mu}^2)\frac{\partial h^{(0)}}{\partial \tilde{\mu}} \right) + \frac{\partial h^{(0)}}{\partial Z^{(0)}}\dot{Z}^{(0)},$$

where $\tilde{\mu} \equiv \cos\zeta$ and $V^* = \frac{1}{2}(V_1^{(0)} + V_2^{(0)}) + (R_1^2 - R_2^2)\dot{Z}^{(0)}/Z^{(0)2}$ is a translational velocity of the origin of the bi-spherical coordinate system in a chosen fixed frame; $c = R_1 \sinh\alpha$ denotes a characteristic length of the bi-spherical coordinate system and $\dot{c} = R_2 \cosh\beta \coth\alpha \dot{Z}^{(0)}/Z^{(0)}$ is its time derivative.

An important result is that the problem for $h^{(1)}$ is quasi-stationary. Its solution depends on time parametrically via the right-hand side of (23), the conditions at infinity (25), and the evolution of the domain. It could be parameterized by Z, since

$$\dot{a}(t) = \frac{\mathrm{d}a}{\mathrm{d}Z}\dot{Z} = \frac{\mathrm{d}a}{\mathrm{d}Z}(V_1^{(0)} - V_2^{(0)}).$$

The solution of (23)–(25) can, thus, be expressed as

$$h^{(1)} = h_0 + h_1 + h_2,$$

where

$$h_1 = -\left(\frac{\mu\dot{a}(t)}{4} + \frac{\mu(1+\mu^2)b(t)}{8}\right)(1 - \mathrm{e}^{-r^2}) \tag{26}$$

satisfies the condition at $|\boldsymbol{x}| \to \infty$,

$$h_2 = -\frac{1}{4\pi}\int_{\mathscr{R}^3} \frac{\Xi^*(\boldsymbol{y})}{|\boldsymbol{x}-\boldsymbol{y}|}\mathrm{d}V(\boldsymbol{y}) \tag{27}$$

satisfies the Poisson equation with the right-hand side $(\Xi - \Delta h_1)$ and decays at infinity. Ξ^* in (27) is a smooth continuation of $(\Xi - \Delta h_1)$ onto $\mathscr{R}^3$. Thus, the function h_0 is harmonic in Ω,

$$\Delta h_0 = 0, \tag{28}$$

and satisfies the boundary conditions

$$h_0 \to 0, \quad |\boldsymbol{x}| \to \infty, \tag{29}$$

$$\frac{\partial h_0}{\partial n} = f(\boldsymbol{x}) = -\frac{\partial h_1}{\partial n} - \frac{\partial h_2}{\partial n}, \quad \boldsymbol{x} \in \partial\Omega. \tag{30}$$

Note that for the equal-sized bubbles, $\dot{Z}(0) = V^{(0)} = 0$, hence $\dot{a} = 0$ and also $b = 0$. In this case $h_1 = 0$, and the constructed solution is the same as that provided by the regular perturbation technique. Moreover, if only the finiteness of the temperature disturbance is required instead of the decay at infinity, the solution constructed formally via *regular* expansion carried to $O(\varepsilon^2)$, results in the same temperature gradients on the interfaces and, hence, still correctly predicts the bubbles migration velocities, while it is clearly incorrect as a solution of the problem, since it fails to vanish at infinity. This is qualitatively the same result as in the problem of steady migration of a single bubble in a thermal gradient at small Pe (Subramanian 1981), where the formal regular expansion carried to $O(Pe^2)$, wherein an incorrect result for the temperature is retained, will correctly predict the bubble migration velocity.

Problem (28)–(30) was solved numerically. The details of the solution are given in Appendix B. Once the temperature distribution on the interface is known, the solution of the hydrodynamic problem can be obtained in a straightforward manner (see Appendix A). The calculation shows that the motion of the leading bubble is always enhanced, while the motion of the trailing one is retarded. The relative velocity $V = V_1 - V_2$ of equal-sized bubbles is plotted in figure 3 as a function of the separation distance $Z - R_1 - R_2$. It is seen that the relative velocity of the equal-sized bubbles tends to zero at both large and at small proximities and it has a maximum at $Z \simeq 2.3$. At large separations the relative velocity of the equal-sized bubbles decays as Z^{-2}, while at close proximity the numerical scheme fails due to the slow convergence of the solution series.

The corrections to the individual velocities of unequal bubbles, $V_i^{(1)}$, versus separation distance are given in figures 4(a) and 4(b) for the cases $R = 1.2$ and $R = 0.6$,

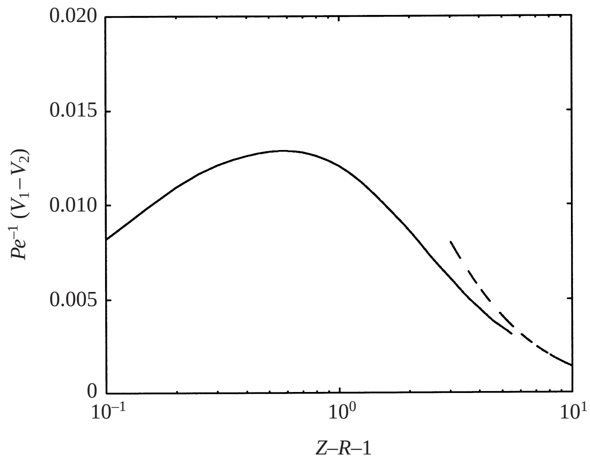

FIGURE 3. The relative velocity of equal-sized bubbles as a function of separation distance. Dashed curve is asymptotic evaluation for large Z (equation (37)).

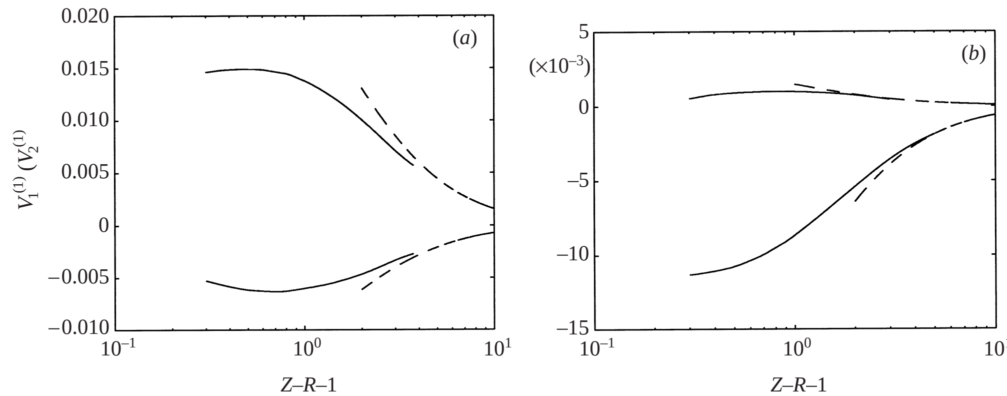

FIGURE 4. The corrections to the individual velocities of unequal bubbles: (*a*) $R = 1.2$ and (*b*) $R = 0.6$. The upper and lower curves correspond to the leading and trailing bubble, respectively. Dashed curves are asymptotic evaluations for large Z (equation (37)).

respectively. It is seen from this figure, that the larger bubble has a more pronounced effect on the smaller one and vice versa. The magnitude of the velocity corrections, $V_i^{(1)}$, decays rapidly with increased separation and thus for large Z the numerical computation is time consuming. An efficient method to study this asymptotic limit is realized in the next subsection.

4.3. *Widely separated bubbles*

When the separation distance is large, $Z \gg R_1 + R_2$, there is another small parameter that enters the problem, $1/Z$. In this sub-section we derive asymptotically valid corrections to the individual migration velocities of widely separated bubbles in closed form.

By making use of the method of reflections (Happel & Brenner 1965; Anderson 1985) the zero-order terms of the inner expansions for $h^{(0)}$ and $\psi^{(0)}$ can be found (see

(C 1)–(C 2) in Appendix C). The individual drift velocities of the bubbles are given by

$$\boldsymbol{V}_i^{(0)} = \frac{R_i}{2}\left[1 + \left(\frac{R_j}{R_i} - 1\right)\frac{R_j^3}{Z^3}\right]\boldsymbol{e}_3 + O(Z^{-5}), \quad i, j = 1, 2, \quad j \neq i. \tag{31}$$

It is readily seen that the solution, constructed in the previous sub-section, is valid for all separations, when $Z \leqslant O(Pe^{-1/2})$, since the ratio of terms neglected in the zero-order outer expansion to those taken into account does not depend on Z. In particular, H_t and U_3 in (16) are of $O(\varepsilon^2/Z^4)$ while $\varepsilon \boldsymbol{U} \cdot \nabla_\rho H$ is of $O(\varepsilon^5/Z^4)$. On the other hand, we know *a priori* that the leading-order correction to the velocities of the bubbles which does not depend on Z is of $O(\varepsilon^4)$ (Subramanian 1981). It follows from (C 1), (C 2) and (31) that for $Z \gg R_1 + R_2$, $H^{(0)}$ is governed by (17) and (18) with

$$\dot{a} = -b = \frac{3R_1^3 R_2^3}{Z^4}(R_1 - R_2). \tag{32}$$

The first-order term of the inner expansion is to be determined from (23)–(25) with $\dot{a}$ and b given by (32). The details of the solution are given in Appendix C. In final form

$$h^{(1)} = h_1^{(1)}(r_1, \theta_1) + h_2^{(1)}(r_2, \theta_2),$$

where

$$h_i^{(1)} = \frac{R_i^4}{12r_i} - \frac{R_i^7}{48r_i^4} + \left(\frac{R_i^6}{9r_i^3} - \frac{R_i^4}{6r_i} - \frac{R_i^7}{24r_i^4}\right)\mathscr{P}_2(\mu_i), \tag{33}$$

(r_i, θ_i, ϕ) denote the local spherical coordinate systems with their origins at the centres of the bubbles with the lines $\theta_i = 0$ parallel to the direction of the applied temperature gradient, $\boldsymbol{e}_3$, and $\mu_i = \cos\theta_i$. Note that $h_i^{(1)}$ coincides with the leading order of the expansion of the temperature field around a single bubble constructed by Subramanian (1981) and Bratukhin (1975). These expansions do not contain the first harmonic and, hence, do not contribute to the translational velocity of the ith bubble. However, this field decays slowly as $1/r_i$ far from the ith bubble and, thus, induces a perturbation of the temperature of $O(Z^{-1})$ in the vicinity of the neighbouring bubble. Moreover, the $\mathscr{P}_2$-term in (33) causes a thermocapillary flow in the vicinity of the ith bubble that is expected to advect the other bubble with some non-zero speed. We next proceed to an accurate evaluation of these perturbations and the induced contribution to the migration velocities.

The Poisson equation (23) is satisfied by $h^{(1)}$ in Ω with the accuracy of $O(Z^{-3})$. Each of the terms $h_i^{(1)}$ in (33) satisfies the boundary conditions on the interface of the ith bubble with the accuracy of $O(Z^{-3})$, while on the interface of the other bubble its normal derivative is of $O(Z^{-1})$. Following the method of reflection we add harmonic functions h_i^* with $\partial h_i^*/\partial n = -\partial h_j/\partial n$, $\boldsymbol{x} \in \Gamma_i$. The constructed solution is of the form

$$h^{(1)} = \sum_{\substack{i=1 \\ j\neq i}}^{2} \frac{R_i^4}{12r_i} - \frac{R_i^7}{48r_i^4} + (-1)^j \frac{R_j^4 R_i^3}{24Z^2 r_i^2}\mathscr{P}_1(\mu_i) + \left(\frac{R_i^6}{9r_i^3} - \frac{R_i^4}{6r_i} - \frac{R_i^7}{24r_i^4}\right)\mathscr{P}_2(\mu_i) + O(Z^{-3}). \tag{34}$$

The correction to the temperature distribution on the interface Γ_i is

$$h^{(1)} = \frac{3R_i^3}{48} - \frac{R_i^4}{12Z} + (-1)^j \frac{R_i R_j^4}{8Z^2}\mathscr{P}_1(\mu_i) - \frac{7R_i^3}{72}\mathscr{P}_2(\mu_i) + O(Z^{-3}). \tag{35}$$

The $\mathscr{P}_2$-term in the temperature distribution (35) generates thermocapillary flow in the vicinity of ith bubble with a stream function

$$\tilde{\psi}_i^{(1)} = \frac{7}{120} R_i^5 \left(\frac{R_i^2}{r_i^2} - 1 \right) \mathscr{C}_3^{-1/2}(\mu_i), \tag{36}$$

which, in turn, translates the jth bubble at a rate $\delta V_{ij}^{(1)}$ due to the reflection of the velocity field from ith bubble. This contribution is found by use of Faxén's theorem as modified for fluid particles (Anderson, 1985). Finally, the corrections to the migration velocity of the bubbles are found, applying the force free condition

$$\begin{aligned} V_i^{(1)} &= \frac{1}{2} \int_{-1}^{1} h_i^{(1)}|_{r_i=R_i} \mathscr{P}_1(\mu_i) d\mu_i + \delta V_{ji}^{(1)} \\ &= (-1)^j \frac{R_j^5}{24Z^2} \left(\frac{R_i}{R_j} + \frac{7}{5} \right) + O(Z^{-3}), \quad i,j = 1,2, \quad i \neq j. \end{aligned} \tag{37}$$

These velocities are depicted versus the separation distance $Z - R_1 - R_2$ in figures $3(a,b)$ and $4(a,b)$ (dashed lines).

Thus, we have found that for widely separated bubbles the influence of the convective transport in their vicinity dominates over the influence of the outer region. This means that in the first approximation in $1/Z$ a regular perturbation applies for all radius ratios and not for equal-sized bubbles only, as for moderate separations, and the singularity appears only at $O(Z^{-4})$.

5. The relative motion of the bubbles

The relative velocity of the bubbles, $V = V_1 - V_2$, can be expanded as

$$V = V^{(0)}(Z,R) + PeV^{(1)}(Z,R) + O(Pe^2), \tag{38}$$

where $V^{(0)}$ is a quasi-stationary relative velocity (Meyyappan *et al.* 1983). As mentioned in §4, $V^{(0)}(Z,R)$ is positive if $R < 1$, i.e. the small bubble is the trailing one. It is negative if $R > 1$ and $V^{(0)}(Z,1) = 0$.

It is readily seen from (38) that, if the bubble sizes are substantially different, the first term in the relative velocity expansion prevails, and the constructed $O(Pe)$ term provides only a small correction to the quasi-stationary velocity. In contrast to this, for equal-sized bubbles the first term vanishes and the second term becomes the leading one. Our results show that $V^{(1)}$ is always positive, hence in this case $V > 0$, and the separation distance grows with time. For unequal bubbles with radius ratio close to one the two leading terms may be of similar magnitude. This case is considered below in more detail.

Let

$$B(Z) = -\frac{\partial V^{(0)}(Z,R)}{\partial R}\bigg| R = 1.$$

It follows from the analysis of the quasi-steady problem (see Appendix A), that $B(Z)$ is positive. The expansion in (38) can be re-written as

$$V = Pe \left(V^{(1)}(Z,1) - \frac{R-1}{Pe} B(Z) \right) + O(Pe^2, (R-1)^2, Pe(R-1)). \tag{39}$$

It is obvious that if

$$Q = \frac{R-1}{Pe} < \frac{V^{(1)}(Z,1)}{B(Z)} = G(Z), \tag{40}$$

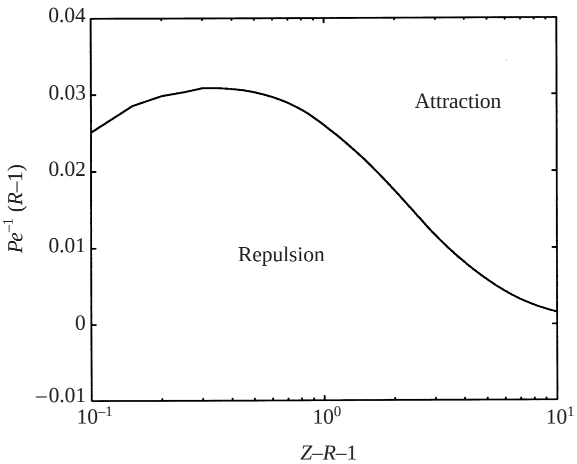

FIGURE 5. Domains of attraction and repulsion of slightly unequal bubbles in the plane of parameters (Z, Q) (equation (40)).

the relative velocity is positive, and the bubbles are repulsed. Note that this condition is always satisfied if $R \leqslant 1$. In the opposite case the relative velocity is negative, and the bubbles are attracted. The domains of attraction and repulsion in the plane of parameters (Z, Q) are shown in figure 5. The function $G(Z)$ has a maximum $G(Z_*) = Q_* \simeq 0.0309$ at $Z_* \simeq 2.3$. For large Z it decays as $0.4Z^{-2}$ as is evident from equations (31) and (37).

Recall now that, while the parameters R and Pe are constants, Z depends on time. Thus, a point in the (Z, Q)-plane which is initially at Z_0 moves parallel to the Z-axis. There are three distinct cases. First if $R \leqslant 1$ then $Q \leqslant 0$, and the system remains in the repulsion domain for all values of Z. The bubbles move apart from each other, and the separation distance grows with time. For large separations the dynamics of this growth can be estimated as follows. If $Q < 0$ $(R < 1)$, $V^{(0)}$ is a positive constant and $V^{(1)}$ decays as $1/Z^2$. Hence, at large Z, the relative velocity is determined by the constant $V^{(0)}$, and the separation distance grows linearly with time. If $Q = 0$ $(R = 1)$ the first term in (39) vanishes and the relative velocity is determined by the second one. An integration of the equation

$$\frac{\mathrm{d}Z}{\mathrm{d}t} = V(Z) \simeq PeV^{(1)} \simeq \frac{0.4Pe}{Z^2}$$

yields that the separation distance grows as $(Pe\,t)^{1/3}$ for $t \to \infty$.

In the second case, if $Q > Q_*$ $(R > 1 + Q_*Pe)$, the point $(Z(t), Q)$ remains in the attraction domain for all t. The bubbles are attracted and eventually collide.

For the third case, consider the situation where $0 < Q < Q_*$ $(1 < R < 1 + Q_*Pe)$. If $Q = G(Z_0)$ the relative velocity equals zero, and the separation distance does not change with time. The unequal bubbles migrate with equal constant velocity, and the process is stationary. If $Q > G(Z_0)$ and the initial separation distance is large enough, $Z_0 > Z_*$, the bubbles are attracted. The separation distance decreases until the point $(Z(t), Q)$ approaches the curve $Q = G(Z)$, where the relative velocity vanishes. After that, the bubbles translate with equal constant velocities keeping a constant separation distance. If $Q > G(Z_0)$ and the initial separation distance is small enough, $Z_0 < Z_*$, the bubbles are attracted and eventually collide as happens for $Q > Q_*$. If $Q < G(Z_0)$

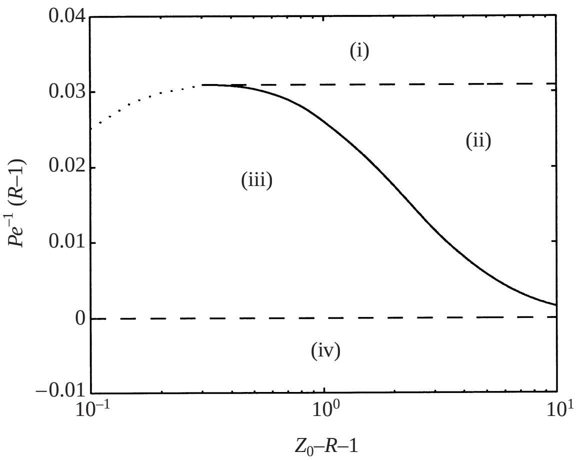

FIGURE 6. Regions of the long-time asymptotic behaviour of slightly unequal bubbles in the space of parameters (Z_0, Q), where Z_0 denotes an initial separation distance: (i) attraction up to collision; (ii) attraction up to migration with equal velocities; (iii) repulsion up to migration with equal velocities; (iv) infinite growth of separation distance.

the bubbles are repulsed. The separation distance grows with time, and the point $(Z(t), Q)$ approaches again the curve $Q = G(Z)$.

Note that for any value of $Q \in (0, Q_*)$ there corresponds one or two values of steady separation distance. It appears that, in contrast to the quasi-stationary situation where the relative velocity between unequal bubbles is always non-zero, the weak convective transport may lead to a selection of a steady separation distance corresponding to the points on the curve $Q = G(Z)$. It follows from the above considerations that for $Z > Z_*$ these steady configurations are stable with respect to the separation perturbations, while for $Z \leqslant Z_*$ they are unstable. Thus, three different long-time asymptotic behaviours of the bubbles motion exist: infinite growth of separation distance, attraction up to collision and attraction or repulsion up to a steady migration of separated bubbles. These are shown in the space of parameters (Z_0, Q) in figure 6.

6. Combined effect of gravity and thermocapillarity

When gravity is not negligible, the momentum equation (2) and the force balance (8) should be modified as follows:

$$Re\,(\partial \boldsymbol{v}_i/\partial t + \boldsymbol{v}_i \cdot \nabla \boldsymbol{v}_i) = -\nabla p_i + \Delta \boldsymbol{v}_i + m\boldsymbol{e}_g, \tag{41}$$

$$\boldsymbol{F}_i = \oint_{\partial\Omega_i} \boldsymbol{\Pi} \cdot \boldsymbol{n}\,\mathrm{d}s - \tfrac{4}{3}\pi R_i^3 m \boldsymbol{e}_g = 0, \quad i = 1, 2, \tag{42}$$

where the dimensionless body force density is $m = (u\eta)^{-1} a_1^2 \rho_o |\boldsymbol{g}|$ with $\boldsymbol{g}$ being the gravitational acceleration and $\boldsymbol{e}_g = \boldsymbol{g}/|\boldsymbol{g}|$. We also assume that the Bond numbers $Bo_i = |\boldsymbol{g}|\rho_o a_i/\sigma_0$, $i = 1, 2$, are small so that the bubbles preserve their spherical shape. The relative influence of gravity and thermocapillary forces on the motion of the bubbles is controlled by m. If $m \leqslant O(1)$ then thermocapillary forces have a considerable effect on the fluid motion; if $m \gg 1$ then the Marangoni effect provides only a small perturbation to the gravity-induced migration of the ith bubble. In what follows we shall focus on the former case of combined effects of gravity and thermocapillarity ($m \leqslant O(1)$) where, for simplicity, $\boldsymbol{e}_g$ is directed parallel to $\boldsymbol{e}_3$ along the line of centres.

Due to the linearity of the quasi-steady problem the zero-order solution can be constructed as a superposition of the solutions of two separate problems: thermocapillary-induced bubble migration and gravity-induced bubble migration, i.e.

$$\psi^{(0)} = \psi_T^{(0)} + \psi_G^{(0)}.$$

The latter problem was studied by Haber *et al.* (1973). It can be shown that far from the bubbles the stream function has the following asymptotic form:

$$\psi^{(0)} \simeq \psi_G^{(0)} = -m\, d(Z,R)(1-\mu^2)\, r + O(1), \tag{43}$$

while the temperature is given by (12).

As in §4, we construct the first correction term to the quasi-steady solution when $0 < Pe \ll 1$. When gravity is present the regular expansion in powers of Pe fails as in §4 due to the slow decay of the velocity field far from the bubbles. Therefore, singular perturbation techniques are used. In what follows we distinguish between two sub-cases: $m \ll 1$ and $m = O(1)$.

6.1. *Weak gravity field, $m \ll 1$*

Following the same arguments as in §4 we let $\varepsilon = \sqrt{Pe}$, and expand H as $H = f_0(\varepsilon)H^{(0)} + f_1(\varepsilon)H^{(1)} + o(f_1(\varepsilon))$, and substitute in (16) together with (43). This defines the following problem for the zero-order term of the outer expansion:

$$f_0(\varepsilon)\Delta_\rho H^{(0)} = f_0(\varepsilon)H_t^{(0)} + U_3^{(0)}(t,\rho,\mu), \tag{44}$$

where

$$U_3^{(0)} = \varepsilon^2 b(t)\mu P_2(\mu)/\rho^2 + \varepsilon m\, d(t)(1+\mu^2)/\rho + O(\varepsilon^3/\rho^3, \varepsilon^2 m/\rho^2). \tag{45}$$

The boundary conditions are as in (18),

$$\lim_{\rho\to\infty} H^{(0)} = 0, \quad \lim_{\rho\to 0} f_0(\varepsilon)\rho^2 H^{(0)}/\varepsilon^2 = \lim_{r\to\infty} r^2 h^{(0)} = a(t)\mu/2. \tag{46}$$

It is readily seen that $f_0(\varepsilon) = \varepsilon^2$ and m is rescaled as $m = \varepsilon\bar{m}$, where $\bar{m} = O(1)$. Following the procedure used in §4.1 we obtain the limit of the outer solution near $\rho = 0$ in terms of the inner spatial variable

$$H = \frac{\mu a(t)}{2r^2} - \varepsilon^2\left(\frac{\mu\dot{a}(t)}{4} + \frac{\mu(1+\mu^2)b(t)}{8} + \frac{4\bar{m}}{3\sqrt{\pi}}\int_0^t \frac{d(\lambda)}{(t-\lambda)^{1/2}}\mathrm{d}\lambda\right) + O(\varepsilon^3). \tag{47}$$

The form of (47) suggests that the leading correction to the temperature inner expansion, $h^{(1)}$, is of $O(\varepsilon^2)$ and, thus, (25) becomes the modified expression

$$\lim_{|\boldsymbol{x}|\to\infty} h^{(1)} = -\left(\frac{\mu\dot{a}(t)}{4} + \frac{\mu(1+\mu^2)b(t)}{8} + \frac{4\bar{m}}{3\sqrt{\pi}}\int_0^t \frac{d(\lambda)}{(t-\lambda)^{1/2}}\mathrm{d}\lambda\right). \tag{48}$$

The addition of the 'history' integral term in (48), being a function of t only, does not contribute to the temperature gradient on the bubble surface at $O(Pe)$. It follows from (48) that the weak gravity field does not change the correction to the bubble migration velocities in the leading order. Thus when $m \leqslant \sqrt{Pe}$ the effect of gravity does not enter the first-order correction of the velocities and is restricted to the temperature field only.

6.2. *Gravity and thermocapillarity of the same order of magnitude,* $m = O(1)$

As in § 6.1 we expand $H(t, \rho, \mu)$ as $H = f_0(\varepsilon)H^{(0)} + f_1(\varepsilon)H^{(1)} + o(f_1(\varepsilon))$ and assume that $\varepsilon = \sqrt{Pe}$. Since m is of $O(1)$, U_3 is of $\mathcal{O}(\varepsilon)$ in the leading order. Therefore, (16) suggests that $f_0(\varepsilon) = \varepsilon$ so that $\varepsilon H_t^{(0)}$ balances $U_3^{(0)}$ Otherwise, (16) would not have solutions that are bounded at infinity. The zero-order term in the outer expansion of the temperature satisfies the non-homogeneous heat equation

$$\varepsilon \Delta_\rho H^{(0)} = \varepsilon H_t^{(0)} + U_3^{(0)}(t, \rho, \mu), \tag{49}$$

where

$$U_3^{(0)} = \varepsilon m\, d(t)(1 + \mu^2)/\rho + O(\varepsilon^2/\rho^2). \tag{50}$$

When $m = O(1)$ the advection in the outer region is governed by the gravity only as a result of slow decay of the velocity far from the bubbles. The corresponding boundary conditions are now

$$\lim_{\rho \to \infty} H^{(0)} = 0, \quad \lim_{\rho \to 0} \rho H^{(0)} = \lim_{r \to \infty} r h^{(0)} = 0. \tag{51}$$

The solution to (49)–(51) can be obtained readily as in § 4.1. Expanding $H^{(0)}$ near $\rho = 0$ up to $O(\varepsilon)$ we obtain

$$H = -\varepsilon \frac{4m}{3\sqrt{\pi}} \int_0^t \frac{d(\lambda)}{(t - \lambda)^{1/2}} \mathrm{d}\lambda + O(\varepsilon^2). \tag{52}$$

This expression indicates that the leading correction to the temperature in the inner region is of $O(\varepsilon)$. Thus, $h^{(1)}$ is a harmonic function satisfying homogeneous Neumann boundary conditions on the bubble surface with the following asymptotic behaviour:

$$\lim_{|\boldsymbol{x}| \to \infty} h^{(1)} = -\frac{4m}{3\sqrt{\pi}} \int_0^t \frac{d(\lambda)}{(t - \lambda)^{1/2}} \mathrm{d}\lambda.$$

The inner expansion of the temperature, $h(t, r, \mu)$, can be constructed as

$$h = h^{(0)} - Pe^{1/2} \frac{4m}{3\sqrt{\pi}} \int_0^t \frac{d(\lambda)}{(t - \lambda)^{1/2}} \mathrm{d}\lambda + O(Pe). \tag{53}$$

It follows from (53) that the integral 'history' term contributes to the uniform heating (cooling) of the fluid in the inner region.

It should be noted that the above $O(Pe^{1/2})$ correction to the temperature also persists in the case of an isolated bubble, when $d = 1$. It appeared impossible to find a steady correction to the temperature field, bounded at infinity, when gravity is included. Thus, the problem is essentially non-steady. In contrast to this, in the case of pure thermocapillary motion (Subramanian 1981), it is justified to consider a steady correction to the temperature field since there is no stokeslet term in the velocity field around the bubble and the convective transport can be balanced by stationary diffusion. On physical grounds, this approach is valid, since the heat is not advected by the thermocapillary motion of the fluid far from the bubble. On the other hand, a gas bubble rising in a liquid with a vertical linear temperature gradient in the presence of gravity, would drag a relatively cold portion of liquid with it towards the hotter region while the diffusion is not fast enough at this order to balance the convective transport. It can be expected that if a perturbation of the Stokesian velocity field is considered simultaneously with the perturbed temperature field, the exponential decay of the velocity perturbations in the outer Oseen region will give rise to stationary temperature corrections.

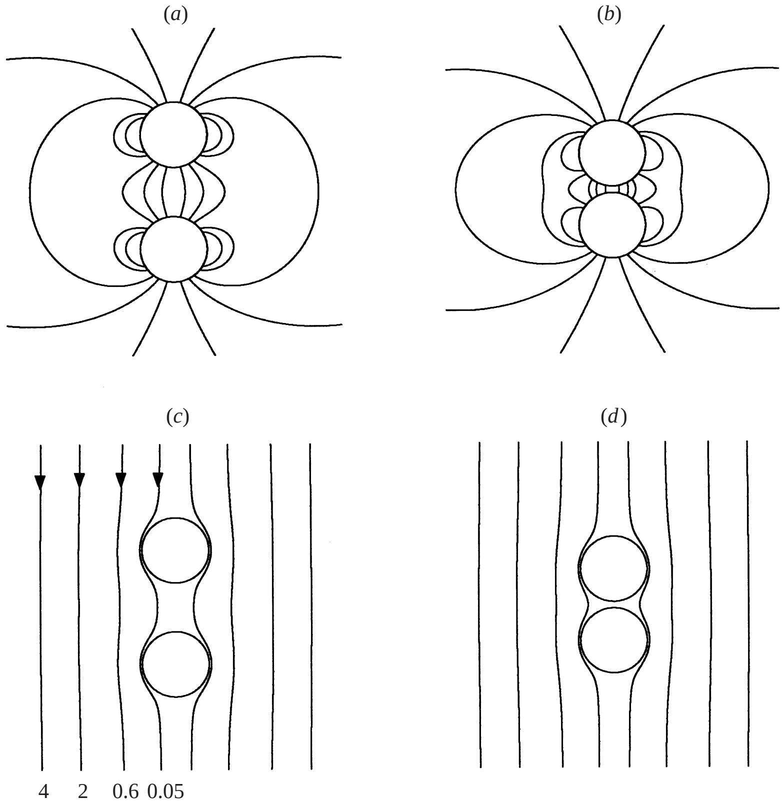

FIGURE 7. Streamlines for the system of equal-sized bubbles at $Pe = 0$: (*a*) $Z - R - 1 = 1.5$, (*b*) $Z - R - 1 = 0.2$ in the laboratory frame; (*c*) $Z - R - 1 = 1.5$, (*d*) $Z - R - 1 = 0.2$ in a frame moving with the bubbles. The streamlines corresponding to the same values of the stream function, $\psi^{(0)}$, are plotted in (*a*) and (*b*), and in (*c*) and (*d*). The numbers denote values of $\psi^{(0)}$ in (*c*) and (*d*).

Finally, the leading-order correction to the bubble migration velocity, due to the influence of gravity, is again of $O(Pe)$ since the constructed integral 'history' term does not alter surface temperature gradients. The corrections to the bubble velocities depend on the next term of the outer temperature expansion, $H^{(1)}$, which has to be computed to determine the asymptotically valid matching condition for the corresponding inner problem for $h^{(2)}$.

7. Discussion

Our results show that, when two bubbles interact, a weak convective transport speeds up the motion of the leading bubble and slows down the motion of the trailing one compared to the migration induced by molecular transport only. To understand this result, recall that for the case of small Pe, the leading-order convective effects are due to the heat transport along the streamlines of the quasi-stationary zero-Pe flow. The characteristic picture of these streamlines for equal-sized bubbles is shown in figure 7(a, b) in a laboratory frame and in figure 7(c, d) in a frame moving with the bubbles. The latter is more useful for understanding the convective effect. When weak convective heat transport takes place, the warmer portion of fluid is brought from infinity to the upstream pole of the leading bubble and also to the downstream pole of the trailing one. The streamline pattern in figure 7(c,d) reveals that a stagnation zone exists in the interparticle region, where the motion of fluid is weak and thus diffusive transport dominates over convection even at finite Pe. This means that the temperature distribution within the interparticle region is not influenced by convective transport as much as that outside this region and the temperature drop over the surface of the leading and trailing bubbles increases and diminishes, respectively. Thus, the leading bubble speeds up while the motion of the trailing one is retarded by thermocapillary convection. In the absence of gravity, the thermocapillary force acting on each bubble is balanced by the viscous resistance so that the particle is force free. At large separation the thermocapillary force exerted on the bubble by the convective transport due to the interaction with the other bubble vanishes. When the bubbles approach each other this force grows, but remains bounded. This behaviour can be understood by viewing, again, figure 7(c,d) where it is evident that the stagnation zone in the interparticle region becomes wider as bubbles approach each other. This by itself would enhance the separation rate. On the other hand the viscous resistance to the relative motion of the bubbles is known to grow without bound as the separation distance vanishes (see e.g. Loewenberg & Davis 1995), so the resulting $V_i^{(1)}$ must then vanish at both $Z \to \infty$ and $Z \to R_1 + R_2$. This means that the corrections to velocities of the bubbles have maxima at some separation distance as a result of an interplay between the thermocapillary and the viscous contributions to the force.

In general, it can be concluded that weak convective transport tends to retard the collision rate of two bubbles. The situation is qualitatively the same as in the case of predominant convective transport analysed by Balasubramaniam & Subramanian (1996). In that case it is due to a different mechanism in which a thermal wake formed past the leading bubble reduces the temperature change over the surface of the trailing one and, thus, substantially retards its thermocapillary-induced motion, while the leading bubble moves with the same speed as when isolated. As a result, the motion of the bubbles towards each other is retarded and the motion away from each other is enhanced. Such behaviour was observed experimentally aboard the space shuttle and was reported by Balasubramaniam *et al.* (1996). In these experiments the Reynolds number was small but the Péclet number was of moderate values. Nevertheless, the qualitative effects on the relative motion of the bubbles in the analysis and in the experiments are remarkably similar.

The results of this paper demonstrate that a weak convective energy transport affects the thermocapillary interaction between two bubbles moving in an external temperature gradient, by adding $O(Pe)$ corrections to the migration velocities of the bubbles. This effect is more pronounced than that obtained by Subramanian (1981, 1983), who found a weaker contribution of $O(Pe^2)$ of the convective transport to the thermocapillary migration velocity of an isolated drop or bubble. In that case, the leading-order perturbation of the temperature field is also of $O(Pe)$, but its influence on the migration velocity vanishes as a consequence of the high symmetry of the flow and temperature fields. Thus, it was necessary to construct higher-order expansion terms in order to obtain the leading-order correction to the migration velocity. This symmetry is broken in the two-bubble migration case.

The perturbations of the migration velocities constructed decay as Pe/Z^2 with the growth of the separation distance. For an isolated bubble the leading-order correction to the migration velocity was found by Subramanian (1981) to be equal to $-\frac{301}{14\,400}Pe^2R_i^3$. Hence, when the bubbles are widely separated, two asymptotic limits can be distinguished: $R_1 + R_2 \ll Z \leqslant Pe^{-1/2}$ and $Pe^{-1/2} \ll Z$. For the first one the $O(Pe)$ correction constructed in the present paper is the leading-order term. For equal-sized bubbles, for which the zero-order migration velocities are identical, it means that the weak convective transport causes the relative motion of the bubbles in the first approximation. In the second case, the $O(Pe^2)$ correction of isolated bubbles is recovered and, hence, in the first approximation the weak convective transport results in retardation of the thermocapillary migration of both bubbles. Note that the $O(Pe)$ correction to the velocity of the smaller bubble is much more pronounced than that to the velocity of the larger one, while the $O(Pe^2)$ term is larger for the larger bubble.

It is interesting to note that in the general case both the temporal change of the mutual positions of the bubble and the convective effects along streamlines contribute to the bubble migration velocities at all separations. This is distinctly different from the case of a spontaneous thermocapillary migration of drops caused by interphase heat or mass transfer (Lavrenteva *et al.* 1999). In that study, the effect of temporal change of the geometry of the flow was found to be more significant than the convective transport along the streamlines for moderate separations and the correction terms obtained are of $O(Pe^{1/2})$. Only when separation is large does the convective transport becomes dominant and the correction reduce to the quasi-steady result of $O(Pe)$. Such a strong difference can be attributed to the fact that, in the case of spontaneous interaction, each drop acts as a source of surfactant with a far concentration field decaying as $1/r$, while in the present case each bubble acts as a thermal dipole, with temperature field decaying as $1/r^2$.

The quasi-steady approximation to the migration velocity of the bubbles ($Pe = 0$) reveals a critical region near $R = 1$ at which the relative motion of the bubbles changes from attraction at $R > 1$ (the leading bubble is smaller) to repulsion at $R < 1$ (the trailing bubble is smaller). The weak convective transport, leading to a small correction to the migration velocities of the bubbles, may result as a major factor influencing the bubble interaction in this critical region. The corrections may change the direction of the relative motion from attraction to repulsion, and lead to the appearance of an interaction pattern that does not exist for $Pe = 0$. For the radii ratio $R = 1 + O(Pe)$ there exist potentially two steady separation distances at which the bubbles migrate with constant velocities. The steady state with the larger separation is stable, while that with the smaller separation is unstable to axisymmetric distortions of the bubble positions. Thus, three different long-time asymptotic behaviours of interacting bubbles are possible: attraction up to collision, infinite repulsion and a stationary translation with equal velocities at some proximity selected by the thermocapillary convection. Note that, although this result was obtained asymptotically assuming small Pe, it is natural to anticipate that such an interaction pattern may also exist for moderate Pe and significantly unequal bubble sizes.

When a constant buoyancy force is included, the stokeslet term in the velocity field leads to a correction to the temperature field in the form of a 'history' term. This term is a spatially homogeneous function of time only and thus does not contribute to the bubble migration velocities at $O(mPe^{1/2})$, where m is the parameter characterizing the relative influence of gravity and thermocapillarity in (42). When gravity forces are weak compared to thermocapillary ones, $m \leqslant Pe^{1/2}$, the results obtained in §§ 4 and 5 for the case of a pure thermocapillary motion are still valid at $O(Pe)$. When these forces are of the same order of magnitude, $m = O(1)$, the leading term of the bubble velocities correction remains of $O(Pe)$; however, its explicit evaluation requires further expansion of the temperature field.

When gravity is negligible, the analysis is expected to be applicable to more general cases: (i) drops with material properties compatible to that of the ambient fluid; (ii) arbitrary orientation of the line of centres with respect to the temperature gradient; (iii) complex systems containing more than two drops or drops adjacent to walls and interfaces. The expected result is that, in these more general cases, the leading-order perturbations of the migration velocity will be of $O(Pe)$ for moderate separation distance.

This work was supported by the Israel Science Foundation. The authors would like to thank Professor H. A. Stone for helpful discussions of this manuscript. O. M. L. acknowledges the support of the Israel Ministry for Immigrant Absorption.

Appendix A. Quasi-stationary solution revisited

A.1. *Hydrodynamic problem*

The general solution of the Stokes equations in Ω, decaying at infinity, can be found using a bi-spherical coordinate system, (ξ, ζ, ϕ), related to the cylindrical coordinate system by $z = c \sinh \xi/(\cosh \xi - \cos \zeta)$, $\omega = c \sin \zeta/(\cosh \xi - \cos \zeta)$. The surface of the first bubble is described by $\xi = \alpha > 0$ and the surface of the second bubble corresponds to $\xi = -\beta < 0$; c is given by $c = R_1 \sinh \alpha = R_2 \sinh \beta$. Note, that R_i is equal to either R (scaled radius of the trailing bubble) or 1. For a given separation distance between the bubble centres, $Z = Z_1 - Z_2$,

$$\cosh \alpha = (Z^2 + R_1^2 - R_2^2)/2ZR_1, \quad \cosh \beta = (Z^2 - R_1^2 + R_2^2)/2ZR_2. \tag{A 1}$$

The stream function ψ has the following form (Stimson & Jeffery 1926; Happel & Brenner 1965; Meyyappan *et al.* 1983)

$$\left.\begin{aligned} \psi &= (\cosh \xi - \tilde{\mu})^{-3/2} \sum_{n=1}^{\infty} W_n(\xi) \mathscr{C}_{n+1}^{-1/2}(\tilde{\mu}), \\ W_n &= a_n^- \cosh n^- \xi + b_n^- \sinh n^- \xi + a_n^+ \cosh n^+ \xi + b_n^+ \sinh n^+ \xi, \end{aligned}\right\} \tag{A 2}$$

where $n^\pm = n + 1/2 \pm 1$, $C_{n+1}^{-1/2}(\tilde{\mu})$ are Gegenbauer polynomials and $\tilde{\mu} \equiv \cos \zeta$; $a_n^\pm$ and $b_n^\pm$ are sets of constants to be determined using the appropriate boundary conditions (4) and (5) for the velocity and temperature fields. The expressions for these constants are (Meyyappan *et al.* 1983)

$$a_n^\pm = \frac{n(n+1)}{2s^\pm}(s_\beta^\pm I_\alpha^n - s_\alpha^\pm I_\beta^n \pm c^2 s_\beta^\pm e_\alpha^\pm V_1 \pm c^2 s_\alpha^\pm e_\beta^\pm V_2),$$

$$b_n^\pm = \mp\frac{n(n+1)}{2s^\pm}(c_\beta^\pm I_\alpha^n + c_\alpha^\pm I_\beta^n - c^2 s_\beta^\pm e_\alpha^\pm V_1 + c^2 c_\alpha^\pm e_\beta^\pm V_2),$$

where

$$(c,s)_{\alpha,\beta}^\pm = (\cosh, \sinh) n^\pm(\alpha, \beta), \quad s^\pm = \sinh n^\pm(\alpha+\beta), \quad e_{\alpha,\beta}^\pm = \frac{\exp(-n^\pm \alpha, \beta)}{\sqrt{2}(n+n^\pm)}.$$

The integrals $I_{\alpha,\beta}^n$ are related to the temperature distribution on the bubble surfaces by

$$I_{\alpha,\beta}^n = -\frac{c^2}{2}\int_{-1}^{1}\left(\frac{\partial \theta}{\partial \tilde{\mu}}\right)_{\xi=\alpha,-\beta}(\cosh(\alpha,\beta) - \tilde{\mu})^{-1/2} \mathscr{C}_{n+1}^{-1/2}(\tilde{\mu})\, \mathrm{d}\tilde{\mu}.$$

Requiring the force acting on each bubble to be zero (Happel & Brenner 1965; Meyyappan *et al.* 1983) gives the following system for the individual bubble velocities

V_1 and V_2:

$$V_1\Delta_1^+ + V_2\Delta_2 = F_\alpha, \tag{A 3}$$

$$V_1\Delta_2 + V_2\Delta_1^- = F_\beta, \tag{A 4}$$

where

$$\Delta_1^\pm = \sum_{n=1}^{\infty} \frac{n(n+1)}{\sqrt{2}(2n-1)(2n+3)}\left[(2n-1)\frac{\exp(\pm n^+(\beta-\alpha))}{s^+} - (2n+3)\frac{\exp(\pm n^-(\beta-\alpha))}{s^-}\right],$$

$$\Delta_2 = \sum_{n=1}^{\infty} \frac{n(n+1)}{\sqrt{2}(2n-1)(2n+3)}\left[(2n+3)\frac{\exp(n^-(\beta+\alpha)}{s^-} - (2n-1)\frac{\exp(n^+(\beta+\alpha))}{s^+}\right],$$

$$F_\alpha = \sum_{n=1}^{\infty}\left(g_\alpha^- I_\beta^n - g_\beta^+ I_\alpha^n\right), \quad F_\beta = \sum_{n=1}^{\infty}\left(g_\alpha^+ I_\beta^n - g_\beta^- I_\alpha^n\right),$$

$$g_{\alpha,\beta}^\pm = \pm\frac{1}{2(2n+1)}\left[\frac{\exp(\pm n^+\alpha,\beta)}{s^+} - \frac{\exp(\pm n^-\alpha,\beta)}{s^-}\right].$$

A.2. *Thermal problem. Zero-order approximation*

Following Sadhal (1983) we introduce the scalar function φ and define the scaled heat flux vector $\boldsymbol{q} = \nabla\theta^{(0)}$ as

$$\boldsymbol{q} = \nabla\theta^{(0)} = \nabla \times \left(\frac{\varphi}{r}\boldsymbol{e}_\phi\right), \tag{A 5}$$

where $\boldsymbol{e}_\phi$ is the unit vector in the ϕ-direction. The bi-spherical components of $\boldsymbol{q}$ are given by

$$q_\xi = \frac{\cosh\xi - \tilde{\mu}}{c}\frac{\partial\theta^{(0)}}{\partial\xi} = -\frac{\cosh\xi - \tilde{\mu}}{cr}\frac{\partial\varphi}{\partial\zeta}, \quad q_\zeta = \frac{\cosh\xi - \tilde{\mu}}{c}\frac{\partial\theta^{(0)}}{\partial\zeta} = \frac{\cosh\xi - \tilde{\mu}}{cr}\frac{\partial\varphi}{\partial\xi}. \tag{A 6}$$

Since $\nabla \times \boldsymbol{q} = 0$, the scalar function φ satisfies $E^2\varphi = 0$ with boundary conditions

$$\varphi = 0, \quad \xi = \alpha, -\beta, \tag{A 7}$$

$$\varphi \to r^2/2, \quad r \to \infty. \tag{A 8}$$

The general solution for the field φ that satisfies (A 8) is

$$\varphi(\xi,\tilde{\mu}) = (\cosh\xi - \tilde{\mu})^{-1/2}\sum_{n=1}^{\infty}(E_n^{(0)}\cosh n_0\xi$$
$$+F_n^{(0)}\sinh n_0\xi + c^2\sqrt{2}n(n+1)\mathrm{e}^{-|\xi|n_0})C_{n+1}^{-1/2}(\tilde{\mu}), \tag{A 9}$$

where $n_0 = n + 1/2$. Using the generating function of Gegenbauer polynomials and (A 7) yields

$$E_n^{(0)} = -\frac{c^2\sqrt{2}n(n+1)}{s^o}(e_\beta^o s_\alpha^o + e_\alpha^o s_\beta^o), \tag{A 10}$$

$$F_n^{(0)} = \frac{c^2\sqrt{2}n(n+1)}{s^o}(e_\beta^o c_\alpha^o - e_\alpha^o c_\beta^o), \tag{A 11}$$

where

$$(c,s)^o_{\alpha,\beta} = (\cosh,\, \sinh) n_0(\alpha,\beta), \quad s^o = \sinh n_0(\alpha+\beta), \quad e^o_{\alpha,\beta} = \exp(-n_0\,\alpha,\beta).$$

It follows from equations (A.1), (A 6) and (A 9) that

$$I^n_{\alpha,\beta} = \mp \frac{c^3\sqrt{2} s^o_{\beta,\alpha}}{s^o}. \tag{A 12}$$

Thus the individual bubble velocities V_1 and V_2 could be found from the set of the two linear equations (A 3), (A 4),

Expanding the formal series solution for the stream function far from the pair of bubbles and taking into account that there is no stokeslet at infinity, one obtains

$$\sum_{n=1}^{\infty}(a_n^- + a_n^+) = 0, \quad b(Z,R_i) = -\sqrt{2}\sum_{n=1}^{\infty}(b_n^- n_- + b_n^+ n_+), \tag{A 13}$$

while $a(Z,R)$ is found from the far field of φ in the same manner,

$$a(Z,R) = -2\sqrt{2}c\sum_{n=1}^{\infty} E_n. \tag{A 14}$$

Appendix B. Numerical procedure

The right-hand side of (30) is computed in a cylindrical coordinate system (ω, z, ϕ). At the point $\boldsymbol{x}$ on the interface with $z = z_x = R_i\cos\theta_i + Z_i$, $\omega = \omega_x = \sqrt{R_i^2 - (z_x - Z_i)^2} = R_i \sin\theta_i$ for $z_x \in (Z_i - R_i, Z_i + R_i)$, $i = 1, 2$.

$$\frac{\partial h_1}{\partial n} = \frac{\omega_x^2 Z_i(1-\mathrm{e}^{-\omega_x^2 - z_x^2})}{8(\omega_x^2+z_x^2)^{3/2}R_i}\left[2\dot{a}(t) + \frac{\omega_x^2+4z_x^2}{\omega_x^2+z_x^2}b(t)\right] - \frac{2(\omega_x^2 + z_x(z_x-Z_i))\,\mathrm{e}^{-\omega_x^2-z_x^2}}{R_i(1-\mathrm{e}^{-\omega_x^2-z_x^2})}h_1(\omega_x, z_x).$$

The normal derivative of h_2 was approximated the second-order central differences while the values of $h_2(\omega, z)$ were calculated from (27) after it was integrated with respect to ϕ:

$$h_2(\omega_x, z_x) = -\frac{1}{\pi}\int_0^{\infty}\int_{-\infty}^{\infty}\frac{\omega \Xi^*(\omega,z)}{[(\omega+\omega_x)^2 + (z-z_x)^2]^{1/2}}\boldsymbol{K}(\boldsymbol{k})\,\mathrm{d}z\mathrm{d}\omega, \tag{B 1}$$

where

$$\boldsymbol{k}^2 = \frac{4\omega\omega_x}{(\omega+\omega_x)^2+(z-z_x)^2},$$

with $\boldsymbol{K}$ being the complete elliptic integral of the first kind with argument $\boldsymbol{k}$, defined as

$$\boldsymbol{K}(\boldsymbol{k}) = \int_0^{\pi/2}\frac{\mathrm{d}\phi}{(1-\boldsymbol{k}^2\cos^2\phi)^{1/2}}.$$

Evaluation of the integral (B 1) was performed numerically. The domain of integration was $\{(\omega, z) \in [0, B] \times [-B, B]\}$, where the value of B was chosen so that the absolute value of the integrand did not exceed 10^{-6} outside this domain.

Once the right-hand side of (30) is computed, the boundary value problem (28)–(30) for h_0 can be solved by making use of the bi-spherical coordinates. The general solution of the Laplace equation in Ω is

$$h_0(\xi,\tilde{\mu}) = (\cosh\xi - \tilde{\mu})^{1/2}\sum_{n=0}^{\infty}\left(E_n^{(1)}\cosh n_0\xi + F_n^{(1)}\sinh n_0\xi\right)\mathscr{P}_n(\tilde{\mu}). \tag{B 2}$$

An application of boundary conditions (30) results in an infinite set of algebraic equations for the coefficients in (B 1):

$$\begin{aligned}(n+1)(E_n^{(1)} - E_{n+1}^{(1)})\sinh n^{+}\xi + n(E_n^{(1)} - E_{n-1}^{(1)})\sinh n^{-}\xi \\ +(n+1)(F_n^{(1)} - F_{n+1}^{(1)})\cosh n^{+}\xi + n(F_n^{(1)} - F_{n-1}^{(1)})\cosh n^{-}\xi \\ = \mp 2n_0 c\int_{-1}^{+1} f(\xi,\tau)(\cosh\xi - \tau)^{-1/2}\mathscr{P}_n(\tau)\,\mathrm{d}\tau, \quad \xi = \alpha, -\beta,\end{aligned} \tag{B 3}$$

where $n_0 = n + 1/2$, $n^{\pm} = n_0 \pm 1$ and f is defined in (30).

The system (B 3) is solved as proposed by Meyyappan *et al.* (1983) by truncating it at some large $n = N$ and setting $E_n = F_n = 0$ for $n > N$.

Appendix C. Widely separated bubbles

Let (r_i, θ_i, ϕ) be local spherical coordinate systems with their origins at the centres of the bubbles and with the lines $\theta_i = 0$ parallel to $\boldsymbol{e}_3$, and let $\boldsymbol{e}_{r_i}$, $\boldsymbol{e}_{\theta_i}$, $\boldsymbol{e}_{\phi}$ be the local basis vectors of these coordinate systems. By making use of the method of reflections (Happel & Brenner 1965; Anderson 1985) the zero-order terms of the inner expansions for $h^{(0)}$ and $\psi^{(0)}$ are found as

$$h^{(0)} = \sum_{\substack{i=1\\ j\neq i}}^{2}\frac{R_i^3}{2r_i^2}\left(1 - \frac{R_j^3}{Z^3}\right)\mathscr{P}_1(\mu_i) + (-1)^j\frac{R_i^5 R_j^3}{r_i^3 Z^4}\mathscr{P}_2(\mu_i) + O(Z^{-5}), \tag{C 1}$$

$$\begin{aligned}\psi^{(0)} = \sum_{\substack{i=1\\ j\neq i}}^{2} -\frac{R_i^4}{2r_i}\left(1 - \frac{R_j^3}{Z^3}\right)\mathscr{C}_2^{-1/2}(\mu_i) \\ +(-1)^j\frac{3R_i^4 R_j^3}{2Z^4}\left(1 - \frac{R_i^2}{r_i^2} - \frac{R_j}{R_i}\right)\mathscr{C}_3^{-1/2}(\mu_i) + O(Z^{-5}),\end{aligned} \tag{C 2}$$

where $\mu_i = \cos\theta_i$.

The terms composing $\Xi\,(\boldsymbol{x}, t)$ in (23) are found from (C 1), (C 2) and (31). For the first one we obtain

$$\begin{aligned}h_t^{(0)} = \sum_{\substack{i=1\\ j\neq i}}^{2}\left(\frac{\partial h^{(0)}}{\partial r_i}\frac{\partial r_i}{\partial t} + \frac{\partial h^{(0)}}{\partial \mu_i}\frac{\partial \mu_i}{\partial t}\right) + \frac{\partial h^{(0)}}{\partial Z}\frac{\mathrm{d}Z}{\mathrm{d}t} = \sum_{\substack{i=1\\ j\neq i}}^{2}\frac{R_i^4}{2r_i^3}\left(1 - \frac{R_j^3}{Z^3}\right)\mathscr{P}_2(\mu_i) \\ +\frac{R_i^7}{r_i^3 Z^3}\left(\frac{R_j}{R_i} - 1\right)\left(\tfrac{4}{3}\mathscr{P}_2(\mu_i) - \tfrac{1}{3}\right) + O(Z^{-4}),\end{aligned} \tag{C 3}$$

where the following equalities have been used:

$$\frac{\partial r_i}{\partial t} = -\mu_i V_i^{(0)}, \quad \frac{\partial \mu_i}{\partial t} = -\frac{(1-\mu_i^2)}{r_i}V_i^{(0)}, \quad \frac{\mathrm{d}Z}{\mathrm{d}t} = V_1^{(0)} - V_2^{(0)}. \tag{C 4}$$

The second term becomes

$$v_3^{(0)} = \sum_{\substack{i=1\\ j\neq i}}^{2} v_{r_i}^{(0)} \cos\theta_i - v_{\theta_i}^{(0)} \sin\theta_i = \sum_{i=1}^{2} \frac{R_i^4}{2r_i^3}\left(1 - \frac{R_j^3}{Z^3}\right)\mathscr{P}_2(\mu_i) + O(Z^{-4}), \tag{C 5}$$

and the last one is of the form

$$\boldsymbol{v}^{(0)}\cdot\nabla h^{(0)} = \sum_{\substack{i=1\\ j\neq i}}^{2}\left(v_{r_i}^{(0)}\frac{\partial h^{(0)}}{\partial r_i} + \frac{v_{\theta_i}^{(0)}}{r_i}\frac{\partial h^{(0)}}{\partial\theta_i}\right) + \left(v_{r_i}^{(0)}\frac{\partial h^{(0)}}{\partial r_j} + \frac{v_{\theta_i}^{(0)}}{r_j}\frac{\partial h^{(0)}}{\partial\theta_j}\right)(\boldsymbol{e}_{r_i},\boldsymbol{e}_{r_j})$$

$$+\left(v_{\theta_i}^{(0)}\frac{\partial h^{(0)}}{\partial r_j} + \frac{v_{r_i}^{(0)}}{r_j}\frac{\partial h^{(0)}}{\partial\theta_j}\right)(\boldsymbol{e}_{r_i},\boldsymbol{e}_{\theta_j}), \tag{C 6}$$

where we made use of symmetry relations

$$(\boldsymbol{e}_{r_i},\boldsymbol{e}_{r_j}) = (\boldsymbol{e}_{\theta_i},\boldsymbol{e}_{\theta_j}), \quad (\boldsymbol{e}_{r_i},\boldsymbol{e}_{\theta_j}) = -(\boldsymbol{e}_{r_j},\boldsymbol{e}_{\theta_i}). \tag{C 7}$$

The term in the first brackets in (C 6), using (C 1) and (C 2), becomes

$$v_{r_i}^{(0)}\frac{\partial h^{(0)}}{\partial r_i} + \frac{v_{\theta_i}^{(0)}}{r_i}\frac{\partial h^{(0)}}{\partial\theta_i} = -\frac{(1+3\mu_i^2)R_i^7}{8r_i^6} + O(Z^{-3}), \tag{C 8}$$

which is uniformly valid in Ω. The second and the third brackets contain the cross-terms which are at least of $O(Z^{-3})$ everywhere in Ω.

Combining these representations we can construct an $O(1)$ solution to (23)–(25) as

$$h^{(1)} = h_1^{(1)}(r_1,\theta_1) + h_2^{(1)}(r_2,\theta_2),$$

where

$$h_i^{(1)} = \frac{R_i^4}{12r_i} - \frac{R_i^7}{48r_i^4} + \left(\frac{R_i^6}{9r_i^3} - \frac{R_i^4}{6r_i} - \frac{R_i^7}{24r_i^4}\right)\mathscr{P}_2(\mu_i). \tag{C 9}$$

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