Thermocapillary Alignment of Gas Bubbles Induced by Convective Transport

Alexander M. Leshansky* and Avinoam Nir†

*Department of Mathematics and †Department of Chemical Engineering, Technion-IIT, Haifa 32000, Israel

Received January 19, 2001; accepted April 21, 2001; published online July 12, 2001

The effect of a weak convective heat transfer on the thermocapillary interaction of two bubbles with an arbitrary orientation relative to an externally imposed temperature gradient is examined. A perturbative analysis of the case of large separation distances, \( Z \), suggests that the corrections to the bubbles' velocities are of \( \mathcal{O}(Pe/Z^2) \), rather than \( \mathcal{O}(Pe^2) \) previously found for an isolated bubble. Equal-sized bubbles are known to move with the same velocities, as if they were isolated, when heat conduction is the only transport mechanism. However, the convective transport results in a relative motion of the bubbles. The tendency of equal bubbles to line up in a plane perpendicular to the applied thermal gradient is shown analytically in the weakly nonlinear limit of small \( Pe \) numbers, and an interesting interaction behavior in the case of unequal bubbles is discussed.

Key Words: thermocapillary migration; hydrodynamic interaction; convective transport; particle alignment.

1. INTRODUCTION

The motion of drops, bubbles, and particles induced by interfacial forces has attracted wide attention in recent years. It is particularly important in cases where the usually dominant buoyancy forces are negligible. If a temperature gradient is applied to a viscous fluid containing gas bubbles with mobile liquid–gas interfaces at zero gravity, the bubbles move toward the hotter fluid owing to the dependence of surface tension on temperature. This effect is most relevant in applications to material processing under conditions of microgravity. Thermocapillary-induced migration of drops and bubbles was first described by Young et al. (1). A review of the subject is given by Subramanian (2) and by Wozniak et al. (3).

Most of the theoretical studies of thermocapillary migration were devoted to the linear limit, i.e., where the effects of convective transport of heat and momentum in the governing equations are completely neglected (negligible Reynolds and Peclet numbers). The surface deformation of spherical drops or bubbles is also usually neglected in the case of small slowly moving particles with high interfacial tension (low capillary numbers).

In this limit the motion is quasi-stationary and the velocities of particles are found as functions of the instantaneous geometrical configuration, the radii ratio, and the material properties of fluids. The axisymmetric case of two bubbles moving along their line of centers in a thermal gradient was treated by Meyyappan et al. (4), who found that the relative velocity of equal-sized bubbles is always zero. Anderson (5) obtained the same result of cancellation of the mutual influence between the two bubbles in the more general case of a thermocapillary migration of two arbitrarily oriented particles with respect to an applied temperature gradient by using the method of reflections. Acivos et al. (6) showed that bubbles exert no influence on each others motion, even when considering a monodispersed system of many interacting bubbles with an arbitrary volume fraction of the bubble phase. Thus, such a bubble system also migrates with equal velocity.

In the nonlinear limit, where the effects of convective transport or inertia become non-negligible, analytical results were reported mainly for the case of an isolated particle. The influence of a weak convective transport, i.e., a small but nonzero Peclet number, on the thermocapillary migration of a single particle was studied by by Bratukhin (7) and by Subramanian (8, 9). The principal result of these studies is that the weak convective transport has only a minor influence on the motion of an isolated drop or bubble. By using singular perturbation techniques it was shown that the corrections to the temperature and velocity fields are proportional to \( Pe \), while the particle migration velocity diminishes by a value of \( O(Pe^2) \) only. Balasubramaniam and Subramanian (10) considered the steady thermocapillary migration of a single bubble at large \( Pe \) and two limiting cases of \( Re = 0 \) and \( Re \to \infty \) and showed that, at both limiting cases, the migration velocity approaches a constant value as \( Pe \to \infty \).

The theory accounting for the convective transport for interacting drops and bubbles is limited. Nas (11) performed a numerical study of interacting drops and bubbles at low and moderate values of \( Re \) and \( Pe \) numbers. He found that two equal bubbles placed initially at arbitrary orientations relative to the applied vertical temperature gradient tend to line up, side by side, perpendicular to this gradient. The simulation of a large monodispersed bubble system showed that the bubbles form a horizontal layer. In polydisperse systems the bubbles of different sizes form different layers, and therefore, a segregation...
by size is observed. Balasubramaniam and Subramanian (12) have studied the migration of two bubbles in a uniform temperature gradient at very large Re and Pe numbers and have shown that the interaction of the trailing bubble with the thermal wake of the leading one results in a substantial retardation of its migration speed. Leshansky et al. (13) reported the influence of weak convective transport on the axisymmetric motion of two gas bubbles in an external temperature gradient. They have found that taking into consideration the week convective transport around the bubbles results in their repulsion with a velocity difference proportional to Pe, which decays like $Z^{-2}$ with the increase of the separation distance, $Z$. Hence, the $O(\text{Pe})$ effect of the convective transport in the case of two bubbles far exceeds the $O(\text{Pe}^2)$ found previously for an isolated bubble.

In the present paper we extend the analysis of Leshansky et al. (13). We consider interacting bubbles moving in an external temperature gradient starting from an arbitrary configuration. The analytically tractable case of distant bubbles is considered. It is expected that this approach may lead to closed form asymptotic expressions for the individual bubble velocities and, thus, will lend the key to a study of the collective effects in dilute dispersions of bubbles. It is predicted that the pronounced convective effect in the case of interacting particles may lead to the nonsymmetric contributions to their migration velocities and, therefore, to a relative motion of the bubbles. It means that, even when they are equal in size, the convective heat transport may result in the breaking of the remarkable result of exact cancellation of the bubble mutual influence, when driven only by conduction (6), and to the interesting behavior of the convection-induced clustering of the bubbles, or the formation of layers as obtained by Nas (11).

2. STATEMENT OF THE PROBLEM

Consider two bubbles of radii $a_1$ and $a_2$ submerged in an unbounded viscous Newtonian fluid that is quiescent at infinity and has a uniform thermal gradient, $T - T_0 = \mathbf{A} \cdot \mathbf{x}$, far away from the bubbles as shown in Fig. 1, with $T_0$ being a reference temperature. The thermal diffusivity, density, and viscosity of the liquid are $\chi$, $\rho$, and $\eta$, respectively. Gravity and buoyancy forces are absent. The viscosity, the density, and the thermal conductivity of the gas phase are assumed to be negligible compared to the same properties in the liquid. Thus, only the governing equations for the liquid need to be solved. It is supposed that the magnitude of $\mathbf{A}a_1$ is smaller than $T_0$ and that 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 changes linearly with temperature: $\sigma = \sigma_0 - \sigma_1(T - T_0)$, where $\sigma_0$ and $\sigma_1$ are positive constants.

The following scaling is chosen: the radius of the larger bubble, $a_1$, for length, $u = \eta_0^{-1} |\sigma| Aa_1$ for velocity, $a_1/u$ for time, $|\sigma| A$ for pressure, and $Aa_1$ for temperature. Let $\mathbf{x} = (x_1, x_2, x_3)$ be a dimensionless radius vector to a point in the laboratory coordinate system with some origin. In this coordinate system let $\mathbf{A} = (0, 0, A)$. Thus, the dimensionless velocity field $\mathbf{v} = (v_1, v_2, v_3)$, the pressure $p$, and the temperature field $\Theta = (T - T_0)/Aa_1$ are described by the following equations and boundary conditions:

$$ Pe(\partial \Theta / \partial t + \mathbf{v} \cdot \nabla \Theta) = \Delta \Theta, \quad \mathbf{x} \in \Omega, \quad [1] $$
$$ Re(\partial \mathbf{v} / \partial t + \mathbf{v} \cdot \nabla \mathbf{v}) = -\nabla p + \Delta \mathbf{v}, \quad [2] $$
$$ \nabla \cdot \mathbf{v} = 0, \quad [3] $$
$$ \Pi \cdot \mathbf{n} = (Ca^{-1} - \Theta) \mathbf{k} \mathbf{n} - \nabla, \quad [4] $$
$$ \mathbf{v} \cdot \mathbf{n} = V_n, \quad [5] $$
$$ \partial \Theta / \partial n = 0, \quad \mathbf{x} \in \Gamma_i \quad [6] $$
$$ \mathbf{v} \to 0, \quad \Theta \to x_3 \text{ at } |\mathbf{x}| \to \infty, \quad [7] $$

where $\Omega$ denotes the domain occupied by the continuous phase, $\Gamma_i$ is the surface of bubble $i$, $V_n$ is the normal interfacial velocity of the $i$th bubble surface, $\Pi = -p \mathbf{I} + (\nabla \mathbf{v} + (\nabla \mathbf{v})^T)$ is the stress tensor, $K = \nabla \cdot \mathbf{n}$ is the curvature of the interface, and $\nabla = \nabla - \mathbf{n} (\mathbf{n} \cdot \nabla)$ denotes the surface gradient. The dimensionless parameters of this problem are the Peclet number $Pe = ua_1/\chi$, the Reynolds number $Re = ua_1\rho/\eta$, and the Capillary number $Ca = |\sigma| Aa_1/\sigma_0$. If $Ca$ is negligible it follows from (4) that the bubbles preserve their spherical shape, $\Omega$: $K = 2, \forall (x \in R^3 : \mathbf{x} - \mathbf{Z}_i(t)) > R_i = a_i/a_1, i = 1, 2). \mathbf{Z}_i(t)$ denotes a radius vector to the center of bubble $i$ at the moment $t$. The total force acting on each bubble is given by

$$ \mathbf{F}_i = \int_{\Gamma_i} \Pi \cdot \mathbf{n} \, ds = 0, \quad i = 1, 2. \quad [8] $$

FIG. 1. Two bubbles migrating in a vertical temperature gradient in an arbitrary orientation. Note that the bubbles’ centers are lying in the plane of the figure, and thus, the vector $\mathbf{Z}$ has no $x_2$ component.
The problem is completed by the kinematic condition
\[
V_i = dZ_i(t)/dt, \quad i = 1, 2, \tag{9}
\]
and by the initial conditions
\[
Z_i(0) = Z_{i0}, \quad i = 1, 2, \tag{10}
\]
\[
\Theta(0, x) = \Theta_0(x), \quad x \in \Omega. \tag{11}
\]

Let the coordinate system be chosen in such a way that the bubbles’ centers lie in the plane of Fig. 1. Symmetry considerations imply that there will be no velocity component in a direction normal to the plane of the figure.

### 3. Construction of the Solution for Small \( Pe \)

The zero-order approximation is the quasi-steady solution of Eqs. (2)–(11) under the following assumptions: \( Re = 0, Ca = 0, \) and \( Pe = 0. \) In this case the thermal and the hydrodynamic problems decouple. The temperature field can be found independently from the velocity field as a harmonic function in \( \Omega \) satisfying the boundary conditions in Eqs. (6) and (7). Once the temperature and the velocity fields are found as solutions of nonevolutionary problems, they depend on time parametrically via the evolution of the problem domain.

The quasi-stationary problem of the motion of two arbitrarily oriented bubbles was first treated by Meyyappan and Subramanian (14) by the use of the approximate technique based on a single reflection of thermal and hydrodynamic disturbances caused by one of the bubbles. They obtained the individual velocities of the bubbles correct to \( O(Z^{-3}) \). Anderson (5) generalized their results for the case of drops and obtained more accurate results correct to \( O(Z^{-6}) \), while Satrape (15) used multipole spherical expansions to solve numerically the same problem for bubbles for all separations. We shall adopt the technique of reflections to construct a zero-order approximation of our problem. The exact solution that can be obtained, in principle, by using the bi-spherical coordinates, however, is cumbersome due to the lack of symmetry present in the axisymmetric problem (Meyyappan et al. (4), Leshansky et al. (13)) and is not expected to alter qualitatively the results of the calculations by simple reflection.

Let \((r_i, \theta_i, \phi_i)\) be the local right-handed spherical coordinate systems with their origins at the centers of the bubbles and with the lines \( \theta_i = 0 \) parallel to \( e_3 \), and let \( e_n, e_0, e_\theta \) be the local basis vectors of these coordinate systems (see Fig. 1). To a first approximation the motion of the two distant bubbles is driven by conduction. Denote the temperature field disturbance by \( h = \Theta - \Theta_0 \) and the stream function by \( \psi \). Then, the solution corresponding to \( Pe = 0 \) (denoted by superscript \((0)\)) may be found via the simple superposition
\[
h_i^{(0)} = \sum_{i=1}^{2} \frac{R_i^3}{2r_i^2} P_i(\mu_i) + O(Z^{-3}), \tag{12}
\]
\[
\psi_i^{(0)} = \sum_{i=1}^{2} \frac{R_i^3}{2r_i^2} C_i^{-1/2}(\mu_i) + O(Z^{-3}), \tag{13}
\]
where \( P_n(\mu_i) \) and \( C_i^{-1/2}(\mu_i) \) denote the Legendre and Gegenbauer polynomials, respectively, \( R_i = a_i/a_1, \) and \( \mu_i \equiv \cos(\theta_i). \) The individual drift velocities of the bubbles are given by
\[
V_i^{(0)} = \frac{R_i}{2} e_3 + O(Z^{-3}), \quad i, j = 1, 2. \tag{14}
\]

The aim of this work is to construct the first correction term to this quasi-stationary solution corresponding to widely separated bubbles when \( 0 < Pe \ll 1. \) Specifically, we are interested in the first correction to the bubbles’ migration velocities, \( V_i = V_i^{(0)}(Z) + \varepsilon(Pe)V_i^{(1)} + O(\varepsilon(Pe)), \) \( \varepsilon(Pe) \ll 1; i = 1, 2. \) Let \( \varepsilon = Pe. \) Thus,
\[
h = h^{(0)} + \varepsilon h^{(1)} + \cdots.
\]

It is clear that for large enough separations the above temperature expansion should match the one found for an isolated bubble,
\[
h_i^{(0)} = \Theta_i^{(0)} + O(Z^{-3}), \quad i, j = 1, 2.
\]

The second term of the temperature disturbance expansion, \( h^{(1)} \), is governed by the boundary value problem
\[
\Delta h^{(1)} = \chi_0(x, t), \quad x \in \Omega, \tag{15}
\]
\[
\partial h^{(1)}/\partial n = 0, \quad x \in \partial \Omega, \tag{16}
\]
\[
\lim_{|x| \to \infty} h^{(1)} = 0, \tag{17}
\]
where
\[
\chi_0(x, t) = h_i^{(0)} + \psi_3^{(0)} + \psi^{(0)} \cdot \nabla h_i^{(0)}.
\]

The terms comprising \( \chi_0(x, t) \) are found from (12)–(14). For the first one we obtain
\[
h_i^{(0)} = \sum_{i=1}^{2} \left( \frac{\partial h_i^{(0)}}{\partial r_i} \frac{\partial r_i}{\partial t} + \frac{\partial h_i^{(0)}}{\partial \mu_i} \frac{\partial \mu_i}{\partial t} \right)
\]
\[
= \sum_{i=1}^{2} \frac{R_i^4}{2r_i^2} P_i(\mu_i) + O(Z^{-3}), \tag{18}
\]
where the following equalities have been used:
\[
\frac{\partial r_i}{\partial t} = -\mu_i V_i^{(0)}(t), \quad \frac{\partial \mu_i}{\partial t} = -\frac{(1 - \mu_i^2)}{r_i} V_i^{(0)}(t). \quad [19]
\]
The second term becomes
\[
v_3^{(0)} = \sum_{i=1}^{2} \frac{R_i^4}{2\mu_i^3} P_2(\mu_i) + O(Z^{-3}),
\]
and the last one is of the form
\[
v^{(0)} \cdot \nabla h^{(0)} = \sum_{i=1}^{2} \left(\frac{v_0^{(0)} \partial h^{(0)}}{r_j} + \frac{v_0^{(0)} \partial h^{(0)}}{\partial \theta_j}\right) (e_r, e_r)\]
\[
= \sum_{i=1}^{2} -\frac{(1 + 3\mu_i^2)R_i^3}{8\mu_i^6} + O(Z^{-3}),
\]
which is uniformly valid in \( \Omega \). This term also contains cross terms, like
\[
\left(v_0^{(0)} \frac{\partial h^{(0)}}{\partial r_j} + v_0^{(0)} \frac{\partial h^{(0)}}{\partial \theta_j}\right) (e_r, e_r)
\]
\[
= \left(v_0^{(0)} \frac{\partial h^{(0)}}{\partial r_j} + v_0^{(0)} \frac{\partial h^{(0)}}{\partial \theta_j}\right) (e_r, e_r),
\]
which are at least of \( O(Z^{-3}) \) everywhere in \( \Omega \). Combining these representations we can construct an \( O(1) \) solution of [15]–[17] as
\[
h^{(1)} = h_1^{(1)}(r_1, \theta_1) + h_2^{(1)}(r_2, \theta_2) + o(Z^{-1}),
\]
where
\[
h_i^{(1)} = \frac{R_i^4}{12r_i} - \frac{R_i^7}{48r_i^4} + \left(\frac{R_i^6}{9r_i^3} - \frac{R_i^4}{6r_i} - \frac{R_i^7}{24r_i^2}\right) P_2(\mu_i). \quad [22]
\]
Note that \( h_i^{(1)} \) coincides with the leading order of the expansion of the temperature field around a single bubble constructed by Bratukhin (7) and Subramaniam (8). These expansions do not contain the first harmonic and, hence, do not contribute to the translational velocity of the bubbles. However, this field decays slowly as \( 1/r_i \) far from the \( i \)th bubble and, thus, induces a perturbation of the temperature of \( o(Z^{-1}) \) in the vicinity of the neighboring bubble. This perturbation is expected to lead to a non-zero contribution to the velocity of the \( j \)th bubble by adding \( \delta V_{ji}^{(1)} \), while the \( \delta P_2 \) term in [22] produces the thermocapillary flow in the vicinity of the \( i \)th bubble that may advect the \( j \)th one at some nonzero rate, \( \delta V_{ji}^{(1)} \). We next proceed to an accurate evaluation of these perturbations and the induced perturbations of the migration velocities.

\( h^{(1)} \) satisfies Poisson’s equation [15] in \( \Omega \) with the accuracy of \( O(Z^{-3}) \). Each of the terms \( h_i^{(1)} \) satisfies the boundary conditions on the interface of the \( i \)th bubble with the accuracy of \( O(Z^{-3}) \), while on the interface of the other bubble its normal derivative is of \( O(Z^{-2}) \). Following the method of reflection, to increase the accuracy of the solution to \( O(Z^{-3}) \), we add harmonic functions \( h_i^{(1)} \) with \( \partial h_i^{(1)}/\partial n = -\partial h_j^{(1)}/\partial n, x \in \Gamma_i \). The constructed solution takes the form
\[
h^{(1)} = \sum_{i=1}^{2} \frac{R_i^4}{12r_i} - \frac{R_i^7}{48r_i^4} + (-1)^j \frac{R_i^4 R_j^3}{24Z^2 r_i^2} F\left(\theta_i, \phi_i, \varphi\right)
\]
\[
+ \left(\frac{R_i^6}{9r_i^3} - \frac{R_i^4}{6r_i} - \frac{R_j^3}{24r_j^2}\right) P_2(\mu_i) + O(Z^{-3}),
\]
where
\[
F(\theta_i, \phi_i, \varphi) = \cos \theta_i \cos \phi_i (9 \cos^2 \varphi - 8)
\]
\[
+ \sin \theta_i \cos \phi_i \sin \phi_i (9 \cos^2 \varphi - 2)
\]
and \( \varphi \) denotes the angle between \( \mathbf{Z} \) and the direction of the temperature gradient at infinity (see Fig. 1). The correction to the temperature distribution on the interface \( \Gamma_i \) becomes
\[
h_i^{(1)} = \frac{3R_i^3}{48} - \frac{R_i^4}{12Z} + (-1)^i \frac{R_i R_j^3}{8Z^2} F(\theta_i, \phi_i, \varphi)
\]
\[
- \frac{7R_i^4}{72} P_2(\mu_i) + O(Z^{-3}). \quad [24]
\]
The contribution to the individual migration velocities of bubbles may be found from [24] using general Lamb’s solution of the Stokes equation in terms of spherical harmonics (see, e.g., Brenner (16)) as
\[
\delta V_{T_{ij}}(t) = \frac{1}{2} \int_{-1}^{1} \int_{-1}^{1} h_i^{(1)}(r_i, \mu_i) P_j(\mu_i) d\mu_i,
\]
\[
\delta V_{T_{ij}}(t) = \frac{1}{4\pi} \int_{0}^{2\pi} \cos \phi_i d\phi_i \int_{-1}^{1} h_i^{(1)}(r_i, \mu_i) P_j(\mu_i) d\mu_i,
\]
where \( P_j(\mu_i) = \sqrt{1 - \mu_i^2} \).

The \( \delta P_2 \) term in the temperature distribution [24] generates an axisymmetric thermocapillary flow in the vicinity of \( i \)th bubble
\[
\tilde{V}_{i}^{(1)} = \frac{7}{120} R_i^5 \left(\frac{R_i^2}{r_i^2} - 1\right) \tilde{\sigma}_3^{-1/2}(\mu_i),
\]
which, in its turn, produces the change in migration velocity of \( j \)th bubble, \( \delta V_{ji}^{(1)} \), due to the reflection of the velocity field generated by \( \tilde{V}_{i}^{(1)} \). These contributions can be determined by the
use of Faxen’s law as modified by Hetroni et al. [17] for fluid particles,

\[
\delta V^{H(1)}_{ji} = \frac{R^2}{2Z^2} \left( \frac{R_j}{2R_j} \cos \varphi (R_j \cos 2\varphi - 7) + \frac{7}{5} \mathcal{P}_2(\cos \varphi) \right)
\]

\[
+ \mathcal{O}(Z^{-3}),
\]

where \( \lambda \) denotes the viscosity ratio and is zero for bubbles. Finally, the combined corrections to the migration velocity of the bubbles are found to be

\[
V_s = \frac{1}{2} Pe \frac{17 \cos 2\varphi - 1}{80Z^2} + \mathcal{O}(Pe^2),
\]

\[
V_s = \frac{1}{2} Pe \frac{\sin 2\varphi}{4Z^2} + \mathcal{O}(Pe^2).
\]

An important observation in Eq. [30] is that the axisymmetric alignment of the bubbles (\( \varphi = 0, \pi \)) is unstable with respect to convection and that any small perturbation in the bubble position will destroy this alignment. On the other hand, the parallel configuration (\( \varphi = \pi/2 \)) is stable, and any small distortion of the bubbles’ position will be suppressed. The pattern of relative motion of two equal-sized bubbles is illustrated in Fig. 2. Analyzing the trajectory pattern, it is seen that the bubbles have a tendency to align in the plane perpendicular to the applied temperature gradient. In the long-time limit any two bubbles would approach each other in the course of this alignment.

Let us now consider two slightly unequal bubbles. In this case the relative velocity of the smaller bubble, 2, with respect to bubble 1, has a contribution from \( V^{(0)} \) and the correction term due to the thermal convection and will take the form

\[
V_r = \frac{\delta}{2} \cos \varphi + Pe \frac{17 \cos 2\varphi - 1}{80Z^2} + \mathcal{O}\left( \frac{\delta}{Z^3}, Pe^2 \right),
\]

\[
V_s = \frac{\delta}{2} \sin \varphi + Pe \frac{\sin 2\varphi}{4Z^2} + \mathcal{O}\left( \frac{\delta}{Z^3}, Pe^2 \right).
\]

where \( \delta = 1 - \alpha_2/\alpha_1 \ll 1 \). The terms proportional to \( \delta \) in [31] and [32] dominate the relative velocity between the bubbles as long as the separation distance, \( Z \), is sufficiently large. The trajectories described by Eq. [30] and illustrated in Fig. 2 will be substantially perturbed in this far region. In the vicinity of the reference bubble the convection-driven terms become comparable to those arising in the leading order (\( Pe = 0 \)). The attraction region that would occupy the whole trajectory space in the case of equal-sized bubbles shrinks to some finite dimensions in the present case of unequal bubbles.

The equations governing the evolution of the mutual positions of the bubbles [31–32] may be rewritten in terms of stretched nondimensional time and distance, \( \tau \) and \( \xi \), respectively,

\[
\frac{d\xi}{d\tau} = \delta^{-1} V_r = -\frac{1}{2} \cos \varphi + \frac{17 \cos 2\varphi - 1}{\xi^2} + \mathcal{O}\left( \frac{1}{\alpha_1^3 \xi^3}, \frac{Pe^3}{\delta} \right).
\]

![FIG. 2. Relative motion of two equal bubbles driven by convective transport in an upward temperature gradient. Arrows correspond to the velocity of a bubble relative to the “reference” bubble (dotted circle) in the coordinate frame moving with the latter. Solid lines represent trajectories of their relative motion.](image)
The motion of a smaller bubble relative to a larger one in a frame moving with the latter, driven by convective transport in an upward temperature gradient. In this figure the interparticle distance is given in units of \( \xi = \left( \frac{2\gamma}{RT} \right)^{1/2} Z \). The dotted circle has radius \( \xi = 1 \). The arrows illustrate the velocity of the smaller bubble, and the solid lines depict the trajectories of its motion. Bold solid lines denote the limiting trajectories. \( a, d, \) and \( c \) denote attraction regions, while \( b \) and \( e \) correspond to the regions of repulsion. Black dots indicate the saddle-point equilibrium positions.

\[
\xi \frac{d\varphi}{d\tau} = \delta^{-1} V_s = \frac{1}{2} \sin \varphi + \frac{20 \sin 2\varphi}{\xi^2} + O\left( \frac{1}{\alpha \xi^2}, \frac{Pe^2}{\delta} \right),
\]

while the variable transform is defined as

\[
Z = \alpha \xi, \quad t = \frac{\alpha}{\delta^{3/2}} \tau, \quad \alpha = \left( \frac{Pe}{80 \delta} \right)^{1/2}.
\]

The usefulness of this similarity rescaling is obvious and is addressed in the discussion section below. The interaction pattern of slightly unequal bubbles is given in Fig. 3. Two distinct equilibrium nodes corresponding to \( V = 0 \) can be calculated for the given value of the parameters \( Pe \) and \( \delta \). The first node relating to an axisymmetric alignment of the bubbles, \( \varphi_* = 0 \) and \( \xi_* = 4\sqrt{2} \approx 5.65 \), was already noted by Leshansky et al. (13). The second node has \( \varphi_* = \cos^{-1}(-3/\sqrt{37}) \approx 2.0866 \) rad and \( \xi_* = 4(15/\sqrt{37})^{1/2} \approx 6.28 \) and corresponds to the asymmetric bubble alignment relative to the external gradient. The latter reveals a ring of equilibrium positions in the physical space. The volume enclosed by the limiting trajectory, which comes from upward infinity and ends at the second equilibrium position (the one separating the regions \( a \) and \( b \) in Fig. 3), and by the limiting trajectory, starting at the trailing pole of the reference bubble and ending at the same equilibrium point (the one separating regions \( e \) and \( c \) in Fig. 3), defines the domain of the bubble attraction. A useful application of the trajectory calculations for two bubbles is the determination of the collision efficiency, \( E_c \), defined as the cross-sectional area enclosed by the figure of revolution formed by the limiting trajectory in upward infinity, normalized by \( \pi (a_1 + a_2)^2 \). If there were no interaction, \( E_c \) would have a value of 1. For the case of two equal bubbles, we obtained \( E_c \approx \infty \). In the case of two slightly unequal bubbles we found that, far from the reference bubble in the direction of upward infinity, the figure of revolution formed by the limiting trajectory forms a cylinder with radius \( \xi \approx 7.042 \) and, thus, the collision efficiency becomes \( E_c \approx 12.399 \alpha^2 = 0.155 \frac{Pe}{\delta} \). The bubbles initially placed outside the region of attraction defined above would miss each other at long times.

4. DISCUSSION

The results of this paper demonstrate that weak convective energy transport affects the thermocapillary interaction between nonconducting gas bubbles, moving in an external temperature gradient. It appeared that when the convective transport is not entirely negligible, \( 0 < Pe \ll 1 \), it adds \( O(Pe/Z^2) \) corrections to the migration velocities of two distant bubbles, with \( Z \) being the separation distance of their centers. These findings are different from those obtained by Subramanian (8, 9), who found that the convective transport has a retardation effect of \( O(Pe^2) \) on the thermocapillary migration of an isolated drop or bubble. It is clear that at some moderate separations, \( Z < Pe^{-1/2} \), the former effect of an interparticle interaction will dominate over the latter effect arising from the convective transport in the vicinity of an isolated bubble. In a general case of unequal bubbles, the constructed perturbations of the migration velocity are small and decay as \( Z^{-2} \) with growth of the separation distance, and thus provide only a small correction to the quasi-steady result corresponding to \( Pe = 0 \). Nevertheless, in the case of a monodispersed suspension of bubbles or when the bubbles are slightly unequal, the weak convective transport dominates the dynamics of the interparticle interaction. Recall that, when the convective transport is entirely neglected, the equal-sized bubbles exert no influence on each other’s velocity for all separation distances (6) due to exact cancellation of the mutual thermal and hydrodynamic disturbances. Hence, the behavior corresponding to \( Pe = 0 \) is not expected to be observed in experimental systems where usually small and moderate values of \( Pe \) are realized (Wozniak et al. (3)).

To understand why the convective heat transport affects the relative motion of the bubbles, let us consider the situation where two bubbles are migrating in a “side-by-side” configuration. Each of the bubbles draws hot fluid along its side. For each bubble the fluid in the interparticle region is pushed down more than on the other side, since the contributions of the two bubbles to the velocity field add in this region. Thus, the temperature in the interparticle region will be higher than outside and the bubbles will be attracted. A different effect is expected in a “tandem” configuration. Here, when weak convective transport takes place, the warmer portion of fluid is brought from infinity...
to the upstream pole of the leading bubble and further to the downstream pole of the trailing one. The velocity of the fluid motion in the interparticle region is small, and the diffusion dominates over the convection even at finite \( Pe \) numbers. It means that the temperature distribution within the interparticle region is not influenced by convective transport as much as that outside this region and that 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 slows down (Leshansky et al. (13)). Since at infinite separation distances the interaction due to convective heat transport is absent and there is no relative motion between equal-sized bubbles, it is obvious that all trajectories must be closed (except for the one on the axis of symmetry). Therefore, any two arbitrarily placed bubbles, even being repulsed initially, would attract each other after some time. Taking into account the symmetries of the trajectory space in the case of equal-sized bubbles, the “four-leafed rose” interaction pattern (see Fig. 2) could be anticipated from purely qualitative arguments.

When the bubbles are slightly unequal \( (\delta = 1 - a_2/a_1 \ll 1) \), it is obvious that at large enough separation distances their relative motion is controlled by heat conduction and the velocity of their relative motion is proportional to \( \delta \), with the larger bubble overtaking the smaller one. At closer separations, \( Z < (Pe/\delta)^{1/2} \), the interaction effect arising due to the convection becomes significant and the relative motion is controlled by the combined action of these two effects (see expressions in Eqs. [33] and [34]). It means that far from the reference bubble all trajectories must be open to infinity, while in its vicinity some of them are closed. We have shown that the rescaled velocity of the relative motion between two slightly unequal bubbles, \( \epsilon^{-1} \mathbf{V} \), depends on two parameters: the stretched separation distance \( Z/\alpha \) where \( \alpha = (Pe/80 \delta)^{1/2} \) and the angle between \( \mathbf{Z} \) and the direction of the temperature gradient, \( \varphi \). Here, \( \alpha \) plays the role of a characteristic length-scale of the interaction distance at which the convection-induced effects balance the conduction-driven motion of the bubbles, while the evolution of their mutual positions occurs on a time scale \( \sim \alpha/\delta^{3/2} \). This rescaling reveals a similarity of the relative motion of two slightly unequal bubbles. Figure 3 illustrates the interaction pattern between slightly unequal bubbles in the stretched coordinates \( \xi \). For any given value of the ratio \( Pe/\delta \) the trajectories in terms of \( Z \) may be produced simply by an appropriate coordinate destretching of the given picture. This pattern may also be produced by perturbing the one given in Fig. 2 by superimposing a constant velocity vector field equal to \(-2\mathbf{e}_x\), at every point in the trajectory space. It is readily seen that trajectory pattern in Fig. 2 is topologically unstable relative to the slight disturbance in \( \delta \) around \( \delta = 0 \).

The union of regions \( a, d \), and \( e \) in Fig. 3 defines the domain of bubble attraction, while the union of \( b \) and \( c \) denotes the repulsion domain. Note that the bubbles belonging to \( a \) would be attracted for every initial orientation, while in \( d \) and \( e \) the bubbles may be initially repulsed before they are attracted in the long-time limit. The equilibrium state lying on the axis of symmetry is a saddle: it is stable to axisymmetric perturbation of the bubble position (the bubble is attracted to this point along the axis \( x_3 \)) while any orbital disturbance relative to the reference bubble will drive the former bubble out of equilibrium. The other equilibrium node lying on the intersection of two limiting trajectories that are open to infinity (Fig. 3) is also a saddle and is thus unstable. Since the collision efficiency \( E_c = 0.155 Pe/\delta \), it is obvious that, if the ratio \( Pe/\delta = O(1) \), the convection-driven interaction between slightly unequal bubbles will be small, and thus, the larger bubble would overtake the smaller one for almost every initial orientation. Therefore, a separation by sizes driven by conduction would be observed, while each size is expected to align horizontally due to the convective heat transport.

To illustrate the tendency of bubble alignment in many particle systems, we computed the trajectories of the relative motion between four equal-sized bubbles induced by the weak convective transport. Only pair interactions between the bubbles were considered at this approximation by integrating Eq. [30] for each bubble pair. This approach is accurate enough when the bubbles are not in a close proximity. The bubbles were initially placed at the vertices of a regular tetrahedron. Two distinct initial
the bubbles are at close proximity, the regular perturbation technique would fail because of a slow decay of the terms $\hat{h}_1^2$, $\psi_1 \sim \frac{1}{\eta}$ in the right-hand side of [15] far from the bubbles, and thus, a matched asymptotic expansion technique must be applied (Subramanian (8), Leshansky et al. (13)). A more accurate calculation of the convection-induced effect is not expected to alter the general qualitative nature of the interaction pattern, since in this system there is no repulsive effect that may oppose the bubble attraction due to the convective heat transport. Note that, although our results were obtained for small $Pe$, it is natural to anticipate that stronger interaction effects may exist for moderate values of $Pe$ numbers, as is supported by numerical results by Nas (11).

Since the problems of thermocapillary-driven migration of bubbles and electrophoretic motion of charged colloidal particles in an external electric field are mathematically similar (see Acrivos et al. (6)), it is anticipated that the effect of the convection-driven clustering of colloidal particles may also occur in the physically different system of particle transport by electrophoresis.

ACKNOWLEDGMENTS

We thank O. M. Lavrenteva and V. Berejnov for the helpful discussions and comments in the course of this investigation. A.M.L. thanks Professor G. Tryggvason for providing the details of the numerical simulations.

REFERENCES