THE AXISYMMETRIC THERMOCAPILLARY MOTION OF TWO FLUID DROPLETS

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Abstract—An exact analytical study is presented for the thermocapillary motion of two spherical droplets in a constant applied temperature gradient along their line of centers. The droplets may be formed from different fluids and have arbitrary radii. The appropriate energy and momentum equations are solved in the quasisteady situation using spherical bipolar coordinates and the droplet velocities are calculated for various cases. The interaction between droplets can be very strong when the surface-to-surface spacing approaches zero. The influence of the interaction, in general, is stronger on the smaller droplet than on the larger one. For the thermocapillary motion of two identical liquid droplets, both migrate faster than the velocity they would possess if isolated. For the specific case of two gas bubbles with equal radii, there is no particle interaction for all separation distances. A comparison between our exact results and predictions from a method of reflections is made. The asymptotic formula for the droplet velocities up to $O(r_{12}^{-6})$, where r_{12} is the center-to-center distance between the droplets, is found to underestimate the effect of droplet interactions; the error can be significant when the droplet surfaces are less than a quarter of the sum of the radii apart.

Key Words: thermocapillary motion, fluid droplet, particle interaction, spherical bipolar coordinates

INTRODUCTION

A droplet of one fluid, when placed in a second, immiscible fluid possessing a temperature gradient, will migrate in the direction of the gradient. This is due to the temperature-induced surface tension gradient at the droplet interface. The thermocapillary migration of droplets was first demonstrated experimentally by Young *et al.* (1959). They also theoretically calculated the migration velocity of a spherical droplet of radius *a* placed in an infinite fluid of viscosity η , with a linear temperature distribution $T_{\infty}(\mathbf{x})$ far away from the droplet. If the droplet is sufficiently small that effects of inertia and convection of energy are negligible, its velocity $U^{(0)}$ is related to the uniform temperature gradient ∇T_{∞} by the following expression:

$$\mathbf{U}^{(0)} = \frac{2}{(2+3\eta^*)(2+k^*)} \frac{a}{\eta} \left(-\frac{\partial\gamma}{\partial T}\right) \nabla T_{\infty}, \qquad [1]$$

where $\partial \gamma / \partial T$ is the gradient of the interfacial tension (γ) with the local temperature (T), and η^* and k^* are the ratios of viscosity and thermal conductivity, respectively, between the internal and surrounding fluid.

Through an exact representation in spherical bipolar coordinates, Meyyappan & Subramanian (1987) solved for the correction to [1] for the quasisteady migration of a gas bubble in the presence of an infinite planar surface due to a uniform temperature gradient which is aligned in an arbitrary direction with respect to the surface. They found that the plane surface exerts the most influence on the bubble when migration occurs normal to it, and the least influence in the case of parallel migration. In general, the boundary effects are relatively weak for the case of thermocapillary migration compared to the case of motion due to a body force such as that caused by gravity.

The axisymmetric problem of two migrating bubbles which are aligned with the undisturbed temperature gradient was solved by Meyyappan *et al.* (1983) using bipolar coordinates, while the case of two arbitrarily oriented bubbles was considered by Meyyappan & Subramanian (1984) using a far-field approximate technique. The more general case of the motion of two arbitrarily oriented fluid droplets due to the Marangoni effect has been analyzed by Anderson (1985) using a method of reflections. Corrections to [1] due to droplet interactions were determined in a power series $1/r_{12}$ up to $O(r_{12}^{-6})$, where r_{12} is the center-to-center distance between the droplets. Anderson

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also used the results for two-droplet interactions to obtain the mean droplet velocity in a bounded suspension to leading order in the droplet volume fraction. An important result of the analyses by Meyyappan *et al.* (1983, 1984) and Anderson (1985) is that the interaction between two droplets is asymptotically of $O(r_{12}^{-3})$ rather than $O(r_{12}^{-1})$, as for the interaction between two Stokeslets (Happel & Brenner 1983); hence, the correction to [1] due to droplet interactions in thermocapillary migration is relatively weak compared to the interaction effects expected in motion driven by gravitational force.

Since the effect of droplet interactions on thermocapillary motion in general is weak unless the droplets are nearly touching, it is particularly important to understand how [1] will be corrected if the gap thickness between the droplets approaches zero. However, when the droplets are close together, the series solution generated from the method of reflections becomes a poor description of droplet interaction effects, due to very low convergence characteristics. In the present work, our objective is to obtain an exact solution to the quasisteady problem of thermocapillary motion of two fluid droplets along their line of centers in the absence of gravity. The droplets may be formed from different fluids and have unequal radii, and the undisturbed temperature gradient is constant over length scales comparable to the system are solved by using the bipolar coordinates and the streamlines for various cases are presented. Our numerical results for the droplet velocities compare favorably with the formulas derived analytically from the method of reflections for the cases of large to moderate particle separations. It is found that the effect of droplet interactions can be severely underestimated by the method of reflections when the droplets are almost in contact.

ANALYSIS

We consider the thermocapillary migration of two spherical droplets of different fluids along their line of centers in a third infinite fluid medium. All of the fluids are assumed to be Newtonian and incompressible. A linear temperature field $T_{\infty}(\mathbf{x})$ with a uniform thermal gradient $E_{\infty}\mathbf{e}_z$ (equal to ∇T_{∞}) is prescribed in the fluid far away from the pair of droplets; \mathbf{e}_z is a unit vector in the cylindrical polar coordinate system (ρ, ϕ, z) . The droplets may differ in radius and in physical properties, but are assumed to maintain their spherical shape. Our purpose here is to determine the correction to [1] for one droplet due to the presence of the other in the temperature and flow fields.

For convenience in satisfying the boundary conditions at droplet interfaces, an orthogonal curvilinear coordinate system (ψ , ζ , ϕ), known as spherical bipolar coordinates (shown in figure 1), is utilized to solve this problem. This coordinate system is related to cylindrical polar coordinates by the following relation in any meridian plane $\phi = \text{const}$ (Morse & Feshbach 1953; Happel & Brenner 1983):

$$\rho = \frac{c \sin \psi}{\cosh \xi - \cos \psi}$$
[2a]

and

$$z = \frac{c \sinh \xi}{\cosh \xi - \cos \psi},$$
 [2b]

where c is a characteristic length in the bipolar coordinate system which is >0. The coordinate surfaces $\xi = \text{const}$ correspond to a family of non-intersecting spheres whose centers lie along the z-axis. Two spheres external to each other are chosen to be $\xi = \xi_1$ (with $\xi_1 > 0$) and $\xi = \xi_2$ (with $\xi_2 < 0$), and the sphere radii a_1 and a_2 as well as the distances of their centers from the origin d_1 and d_2 are given by

$$a_i = c \operatorname{cosech} |\xi_i|$$
^[3]

and

$$d_i = c \, \coth \left| \, \xi_i \right|, \tag{4}$$

for i = 1 or 2. The center-to-center distance between the particles, r_{12} , equals $(d_1 + d_2)$.



Figure 1. Geometric sketch for the motion of two spheres.

To determine the migration velocities of the two droplets, it is necessary to ascertain the temperature and velocity distributions.

Temperature distribution

When the migration velocities of the two droplets are not identical, the transport of momentum and energy is inherently unsteady. However, the problem can be considered quasisteady if Péclet and Reynolds numbers are small. The energy equation governing the temperature distribution $T(\mathbf{x})$ for external fluid of constant thermal conductivity k is Laplace's equation:

$$\nabla^2 T = 0.$$
 [5a]

For the two droplets, one has

$$\nabla^2 T_i = 0, \qquad i = 1 \text{ or } 2, \tag{5b}$$

where $T_1(\mathbf{x})$ and $T_2(\mathbf{x})$ are the temperature fields inside droplets 1 and 2, respectively. The boundary conditions require that the temperature and the normal component of heat flux be continuous at the droplet interface and that the temperature field far away from the droplets approach the undisturbed values. Thus,

$$\xi = \xi_i : \qquad T = T_i, \qquad [6a]$$

$$k\frac{\partial T}{\partial \xi} = k_i \frac{\partial T_i}{\partial \xi},$$
 [6b]

$$(\rho^2 + z^2)^{1/2} \to \infty; \quad T \to T_\infty = E_\infty z, \tag{6c}$$

for i = 1 or 2; k_1 and k_2 are the thermal conductivities of the internal fluids, which are assumed to be independent of temperature.

A general solution to the Laplace equation [5a] suitable for satisfying these boundary conditions is (Morse & Feshbach 1953)

$$T = cE_{\infty}(\cosh \xi - \mu)^{1/2} \sum_{n=0}^{\infty} \left[A_n \cosh(n + \frac{1}{2})\xi + B_n \sinh(n + \frac{1}{2})\xi\right] P_n(\mu) + E_{\infty} z,$$
 [7a]

where P_n is the Legendre polynomial of order *n* and for brevity we have put $\mu = \cos \psi$. Boundary condition [6c] is immediately satisfied by a solution of this form. Because the temperature is finite for any position in the interior of each droplet, the solution to [5b] can be expressed as

$$T_{i} = cE_{\infty}(\cosh \xi - \mu)^{1/2} \sum_{n=0}^{\infty} C_{in} \exp[-(n + \frac{1}{2})|\xi|] P_{n}(\mu) + E_{\infty}z, \qquad [7b]$$

for i = 1 or 2. The coefficients A_n , B_n , C_{1n} and C_{2n} in [7a,b] are to be determined using [6a,b]. Utilizing the expansion, which can be derived using the generating function of the Legendre polynomials,

$$\frac{\cosh \xi}{(\cosh \xi - \mu)^{1/2}} - \frac{\sinh^2 \xi}{(\cosh \xi - \mu)^{3/2}} = \sqrt{2} \sum_{n=0}^{\infty} \exp\left[-(n + \frac{1}{2})\left|\xi\right|\right] \times \left[\cosh \xi - (2n + 1)\sinh\left|\xi\right|\right] P_n(\mu) \quad [8]$$

and the recurrence relations of the Legendre polynomials, one can apply the boundary conditions [6a,b] at the droplet interfaces to the general solution [7a,b] to yield four algebraic recursion formulas, as shown in table 1. In table 1, $k_i^* = k_i/k$ is the ratio of thermal conductivity between the internal and surrounding fluid. The four formulas represent a group of infinite coupled equations for the unknown coefficients A_n , B_n , C_{1n} and C_{2n} . Because these coefficients should individually approach zero as $n \to \infty$ for the temperature field [7a,b] to remain bounded, they can be determined by solving the first m sets of the four recursion equations, provided that m is sufficiently large that all A_{m+1} , B_{m+1} , $C_{1(m+1)}$ and $C_{2(m+1)}$ are negligible.

Fluid velocity distribution

With knowledge of the solution for the temperature field, we can now proceed to find the fluid velocity distribution. Due to the low Reynolds numbers encountered in thermocapillary motions, the fluid velocity inside and outside the droplets is governed by the quasisteady fourth-order differential equation for viscous axisymmetric flows:

$$E^4 \Psi = 0, [9a]$$

$$E^4 \Psi_i = 0, \qquad i = 1 \text{ or } 2,$$
 [9b]

where Ψ_i and Ψ are the Stokes stream functions for the flow inside droplet *i* and for the external flow, respectively. The operator E^2 assumes the following form in spherical bipolar coordinates:

$$E^{2} \equiv \frac{\cosh \xi - \mu}{c^{2}} \left\{ \frac{\partial}{\partial \xi} \left[(\cosh \xi - \mu) \frac{\partial}{\partial \xi} \right] + (1 - \mu^{2}) \frac{\partial}{\partial \mu} \left[(\cosh \xi - \mu) \frac{\partial}{\partial \mu} \right] \right\}.$$
 [10]

Table 1. Recursion formulas for the evaluation of the coefficients in [7a, b] (both formulas are valid for i = 1 or 2)

$C_{in} \exp\left[-(n+\frac{1}{2}) \xi_i \right] = A_n \cosh(n+\frac{1}{2})\xi_i + B_n \sinh(n+\frac{1}{2})\xi_i$
$\frac{(n+1)\left[\sinh(n+\frac{3}{2})\xi_{i}+\frac{\xi_{i}}{ \xi_{i} }k_{i}^{*}\cosh(n+\frac{3}{2})\xi_{i}\right]A_{n+1}+(n+1)\left[\cosh(n+\frac{3}{2})\xi_{i}+k_{i}^{*}\sinh(n+\frac{3}{2}) \xi_{i} \right]B_{n+1}}{(n+1)\left[\cosh(n+\frac{3}{2})\xi_{i}+k_{i}^{*}\sinh(n+\frac{3}{2}) \xi_{i} \right]B_{n+1}}$
$-\left\{(1-k_i^*)\sinh\xi_i\cosh(n+\frac{1}{2})\xi_i+\left[\sinh(n+\frac{1}{2})\xi_i+\frac{\xi_i}{ \xi_i }k_i^*\cosh(n+\frac{1}{2})\xi_i\right](2n+1)\cosh\xi_i\right\}A_n$
$-\left\{(1-k_i^*)\sinh\xi_i\sinh(n+\frac{1}{2})\xi_i+\left[\cosh(n+\frac{1}{2})\xi_i+k_i^*\sinh(n+\frac{1}{2}) \xi_i \right](2n+1)\cosh\xi_i\right\}B_n$
$+ n \left[\sinh(n - \frac{1}{2})\xi_i + \frac{\xi_i}{ \xi_i } k_i^* \cosh(n - \frac{1}{2})\xi_i \right] A_{n-1} + n \left[\cosh(n - \frac{1}{2})\xi_i + k_i^* \sinh(n - \frac{1}{2}) \xi_i \right] B_{n-1}$
$= 2\sqrt{2}(1-k_i^*) [\cosh \xi_i - (2n+1)\sinh \xi_i] \exp[-(n+\frac{1}{2}) \xi_i]$

The stream function Ψ (or Ψ_i) is related to the velocity field v (or v_i) by

$$v_{\xi} = \frac{(\cosh \xi - \cos \psi)^2}{c^2 \sin \psi} \frac{\partial \Psi}{\partial \psi}$$
[11a]

and

$$v_{\psi} = -\frac{(\cosh\xi - \cos\psi)^2}{c^2\sin\psi}\frac{\partial\Psi}{\partial\xi}.$$
 [11b]

The interfacial tension is temperature dependent, so the tangential stress discontinuity at the droplet interfaces is

$$\xi = \xi_i: \quad \nabla_{\mathbf{s}} \gamma_i = (\mathbf{I} - \mathbf{nn}) \cdot \nabla \gamma_i = \frac{\partial \gamma_i}{\partial T} \nabla_{\mathbf{s}} T, \qquad [12]$$

where γ_i is the interfacial tension for the surface of droplet *i*, **n** is the unit normal vector at the droplet surface pointing into the surrounding fluid and **I** is the unit tensor. Note that $\partial \gamma_i / \partial T$ is assumed constant on the scale of radius of droplet *i*, and $\nabla_s T$ can be determined from [7a,b]. The boundary conditions for the velocity fields are:

$$\boldsymbol{\xi} = \boldsymbol{\xi}_i: \qquad \mathbf{v} = \mathbf{v}_i, \qquad [13a]$$

$$\mathbf{e}_{\xi} \cdot (\mathbf{v} - \mathbf{U}_i) = \mathbf{0}, \qquad [13b]$$

$$(\mathbf{I} - \mathbf{e}_{\xi} \mathbf{e}_{\xi}) \mathbf{e}_{\xi} : (\tau - \tau_i) = -\nabla_{\mathbf{s}} \gamma_i, \qquad [13c]$$

$$(\rho^2 + z^2)^{1/2} \to \infty; \qquad \mathbf{v} \to \mathbf{0}, \qquad [13d]$$

for i = 1 or 2. Here, $\tau (=\eta[(\nabla v + (\nabla v)^T])$ and τ_i are viscous stress tensors for the external flow and the flow inside droplet *i*, respectively; \mathbf{e}_{ξ} is a unit vector in bipolar coordinates; and $\mathbf{U}_1 (= U_1 \mathbf{e}_z)$ and $\mathbf{U}_2 (= U_2 \mathbf{e}_z)$ are the instantaneous thermocapillary velocities of the two droplets to be determined.

Because the droplets are freely suspended in the surrounding fluid, the net force exerted by the fluid on the surface of each droplet must vanish:

$$\mathbf{F} = \iint_{\substack{\text{droplet}\\\text{interface}}} \mathbf{n} \cdot \mathbf{\Pi} \, \mathrm{d}S = \mathbf{0}, \qquad [14]$$

where Π is the total stress tensor. For the axisymmetric motion considered in this work, one can evaluate U_1 and U_2 by merely satisfying constraint [14] after solving [9a,b] and [13a-d].

A general solution of [9a,b] satisfying boundary condition [13d] and the requirement of finite velocity in the interior of each droplet is (Stimson & Jeffery 1926; Happel & Brenner 1983)

$$\Psi = c^{2} (\cosh \xi - \mu)^{-3/2} \sum_{n=1}^{\infty} [a_{n} \cosh(n - \frac{1}{2})\xi + b_{n} \sinh(n - \frac{1}{2})\xi + c_{n} \cosh(n + \frac{3}{2})\xi + d_{n} \sinh(n + \frac{3}{2})\xi] G_{n+1}^{-1/2}(\mu)$$
[15a]

and

$$\Psi_{i} = c^{2} (\cosh \xi - \mu)^{-3/2} \sum_{n=1}^{\infty} \left\{ e_{in} \exp[-(n - \frac{1}{2}) |\xi|] + f_{in} \exp[-(n + \frac{3}{2}) |\xi|] \right\} G_{n+1}^{-1/2}(\mu), \quad [15b]$$

for i = 1 or 2. $G_{n+1}^{-1/2}(\mu)$ is the Gegenbauer polynomial of order n + 1 and degree -1/2, which is related to Legendre polynomials by

$$G_{n+1}^{-1/2}(\mu) = \frac{P_{n-1}(\mu) - P_{n+1}(\mu)}{2n+1}.$$
 [16]

The coefficients a_n , b_n , c_n , d_n , e_{in} and f_{in} are to be determined from boundary conditions given by [13a-c] using the recurrence relations of the Legendre polynomials as well as expansions derived from the generating function of the Legendre polynomials. The procedure is straightforward but tedious and the results, which consist of eight algebraic recursion formulas, are listed in table 2. In table 2, $\eta_i^* = \eta_i/\eta$ is the ratio of viscosity between the internal and continuous fluid. Because Table 2. Recursion formulas for the evaluation of the coefficients in [15a,b] (all formulas are valid for i = 1 or 2)

$$\frac{a_n \cosh(n-\frac{1}{2})\xi_i + b_n \sinh(n-\frac{1}{2})\xi_i + c_n \cosh(n+\frac{3}{2})\xi_i + d_n \sinh(n+\frac{3}{2})\xi_i}{= -\frac{\sqrt{2}}{2} U_i n(n+1) \left\{ \frac{\exp[-(n-\frac{1}{2})|\xi_i|]}{2n-1} - \frac{\exp[-(n+\frac{3}{2})|\xi_i|]\right\}}{2n+3} \right\}$$

$$\frac{e_{in} \exp[-(n-\frac{1}{2})|\xi_i|] + f_{in} \exp[-(n+\frac{3}{2})|\xi_i|] = -\frac{\sqrt{2}}{2} U_i n(n+1) \left\{ \frac{\exp[-(n-\frac{1}{2})|\xi_i|]}{2n-1} - \frac{\exp[-(n+\frac{3}{2})|\xi_i|]\right\}}{2n+3} \right\}$$

$$\begin{aligned} &-\frac{\nabla_{i}}{|\xi_{i}|} \{(n-\frac{1}{2})e_{m} \exp[-(n-\frac{1}{2})|\xi_{i}|] + (n+\frac{3}{2})f_{m}^{2} \exp[-(n+\frac{3}{2})|\xi_{i}|]\} \\ &= (n-\frac{1}{2})[a_{n} \sinh(n-\frac{1}{2})\xi_{i} + b_{n} \cosh(n-\frac{1}{2})\xi_{i}] + (n+\frac{3}{2})[c_{n} \sinh(n+\frac{1}{2})\xi_{i}] + (n+\frac{3}{2})[c_{n} \sinh(n+\frac{1}{2})\xi_{i}] \\ &+ (n+\frac{1}{2})^{2}a_{n} \cosh(n-\frac{1}{2})\xi_{i} + b_{n} \sinh(n-\frac{1}{2})\xi_{i}] + (n+\frac{3}{2})^{2}[c_{n} \cosh(n+\frac{1}{2})\xi_{i}] + (n+\frac{3}{2})\xi_{i}] \\ &+ (n+\frac{1}{2})^{2}n + (n+\frac{1}{2})\xi_{i} + b_{n+1} \sinh(n+\frac{1}{2})\xi_{i}] + (n+\frac{1}{2})^{2}[c_{n} \cosh(n+\frac{1}{2})\xi_{i}] + (n+\frac{1}{2})\xi_{i}] \\ &+ (n-\frac{1}{2})^{2}(n+1) [a_{n-1} \cosh(n-\frac{1}{2})\xi_{i} + b_{n-1} \sinh(n-\frac{3}{2})\xi_{i}] + (n+\frac{1}{2})^{2}(n+1) [c_{n-1} \cosh(n+\frac{1}{2})\xi_{i} + d_{n-1} \sinh(n+\frac{1}{2})\xi_{i}] \\ &- \frac{3}{2} \sinh \xi_{i} \{(n-\frac{1}{2})[a_{n} \sinh(n-\frac{1}{2})\xi_{i} + b_{n} \cosh(n-\frac{1}{2})\xi_{i}] + (n+\frac{3}{2})f_{n} \exp[-(n+\frac{3}{2})]\xi_{i}]\} \\ &- \eta^{*} \bigg[- \cosh \xi_{i}^{1}(n-\frac{1}{2})^{2}e_{m} \exp[-(n-\frac{1}{2})]\xi_{i}] + (n+\frac{3}{2})^{2}f_{m} \exp[-(n+\frac{3}{2})]\xi_{i}] \bigg] \\ &+ (n+\frac{1}{2})^{2}n \\ &+ (n+\frac{1}{2})^{2}n \\ &+ (n+\frac{1}{2})^{2}(n+1) \\ 2n-1 e_{n-1} \exp[-(n+\frac{1}{2})]\xi_{i}] + (n+\frac{3}{2})^{2}n \\ &+ (n+\frac{1}{2})^{2}(n+1) \\ &2n-1 \\ &- (n+\frac{1}{2})|\xi_{i}|] + (n+\frac{3}{2})^{2}n \\ &+ (n+\frac{1}{2})^{2}(n+1) \\ &- (n+\frac{1}{2})|\xi_{i}|] + (n+\frac{3}{2})f_{m} \exp[-(n+\frac{3}{2})]\xi_{i}]] \bigg] \\ \\ &+ \frac{\sqrt{2}}{2n} \frac{\sqrt{2}}{n} (n^{*} - 1)U_{i}n(n+1) \cosh \xi_{i} \bigg\{ \exp[-(n-\frac{1}{2})]\xi_{i}] + (n+\frac{3}{2})f_{m} \exp[-(n+\frac{3}{2})]\xi_{i}] \bigg\} \\ &+ \frac{\partial\gamma_{i}}E_{n,c}}{\partial T} \sqrt{2n(n+1)} \frac{\xi_{i}}{|\xi_{i}|} \bigg\{ \exp[-(n-\frac{1}{2})|\xi_{i}|] - \exp[-(n+\frac{1}{2})|\xi_{i}|] \bigg\} \\ &+ \frac{\partial\gamma_{i}}E_{n,c}}{\partial T} \left\{ -\frac{n(n+1)}{(h-1)} \frac{\xi_{i}}{(n+1)} \exp[-(n-\frac{1}{2})|\xi_{i}|] - \frac{n(n+1)(n+2)}{2n+3}} e_{n+1} \exp[-(n+\frac{3}{2})|\xi_{i}|] \bigg\} \\ &+ n(n+1) \cosh \xi_{i}c_{m} \exp[-(n+\frac{1}{2})|\xi_{i}|] - \frac{n(n+1)(n+2)}{2n+3}} e_{n+1} \exp[-(n+\frac{3}{2})|\xi_{i}|] \\ &+ \frac{(n-1)n(n+1)}{2n-1} e_{n-1} \exp[-(n-\frac{1}{2})|\xi_{i}|] \bigg\}$$

the unknown coefficients a_n , b_n , c_n , d_n , e_{1n} , e_{2n} , f_{1n} and f_{2n} become small with large *n*, simultaneous solution of these eight recursion equations for the first *m* sets yields 8*m* coefficients, thereby determining the Stokes stream function for fluids according to [15a,b].

By integration of the stress vector on the outer side of the interface using the first part of [14], the drag force opposing the thermocapillary motion of the droplet at $\xi = \xi_1$ is (Stimson & Jeffery 1926)

$$F_{1} = -2\sqrt{2}\pi\eta c \sum_{n=1}^{\infty} (a_{n} + b_{n} + c_{n} + d_{n}); \qquad [17a]$$

and for the droplet at $\xi = \xi_2$,

$$F_2 = -2\sqrt{2}\pi\eta c \sum_{n=1}^{\infty} (a_n - b_n + c_n - d_n).$$
 [17b]

Equivalent formulas expressed in terms of the coefficients for internal stream functions can be obtained easily.

Droplet velocities

Since the net force acting on each droplet vanishes to fulfill the requirement of [14], we have

$$F_1 = 0$$
 and $F_2 = 0.$ [18a,b]

To determine the instantaneous thermocapillary velocities U_1 and U_2 of the droplets, the above two equations incorporated with [17a, b] must be solved. The result can be expressed as

$$U_1 = M_{11} U_1^{(0)} + M_{12} U_2^{(0)}$$
[19a]

and

$$U_2 = M_{21} U_1^{(0)} + M_{22} U_2^{(0)}, \qquad [19b]$$

where $U_1^{(0)}$ and $U_2^{(0)}$ are the droplet velocities that would exist in the absence of the other and are computed from [1]. The numerical results for the mobility coefficients M_{11} , M_{12} , M_{21} and M_{22} as a function of η_1^* , k_1^* , η_2^* , k_2^* , a_2/a_1 and the separation parameter λ , defined as $(a_1 + a_2)/r_{12}$, have been determined and are presented in the following section. Note that $M_{11} = M_{22} = 1$ and $M_{12} = M_{21} = 0$ when the two droplets are separated by an infinite distance (i.e. $\lambda = 0$).

RESULTS AND DISCUSSION

The coefficients of the temperature distribution [7a,b] and the stream function [15a,b] in the present quasisteady problem have been calculated for various values of η_1^* , η_2^* , k_1^* , k_2^* , a_2/a_1 and λ using a digital computer. For the case of $a_2/a_1 = 5.0$ and $\lambda = 0.995$, *m* equal to about 280 was sufficiently large that the (m + 1)th terms of these coefficients are negligible and an increase in *m* does not alter the calculated values appreciably.

Streamlines

The distortion of the velocity field due to interactions between two fluid droplets in thermocapillary motion is illustrated graphically in figures 2–6. In each case, streamlines in a meridian plane



Figure 2. Streamlines for the system $\eta_1^* = k_1^* = \eta_2^* = k_2^* = 1$, $\partial \gamma_1 / \partial T = \partial \gamma_2 / \partial T$, $a_2 / a_1 = 1.0$ and $\lambda = 0.5$. $\Psi / c^2 U_1^{(0)}$: 1, -0.01; 2, -0.03; 3, -0.06; 4, -0.09; 5, -0.12.



Figure 3. Streamlines for the system $\eta_1^* = k_1^* = k_2^* = 1$, $\eta_2^* = 10$, $\partial \gamma_1 / \partial T = \partial \gamma_2 / \partial T$, $a_2 / a_1 = 1.0$ and $\lambda = 0.5$. $\Psi / c^2 U_1^{(0)}$: 1, -0.0125; 2, -0.0375; 3, -0.05; 4, -0.075; 5, -0.125.



4, -0.4; 5, -0.6.





are depicted. Figures 2-5 show the situation when the two spheres have identical radii and the distance between the interfaces is equal to the sum of their radii. The streamlines for two identical droplets with the same viscosity and thermal conductivity as the surrounding fluid are drawn in figure 2. Similar to that for two equal-sized gas bubbles (Meyyappan *et al.* 1983), the contour pattern shows equivalent local recirculations in the vicinity of each sphere and a global recirculation pattern far away from the droplets. These balanced recirculations will be distorted if the two droplets differ in viscosity, in thermal conductivity or in the variation of interfacial tension with temperature, as shown in figures 3-5. It can be found that the spacing between streamlines in the droplet with smaller viscosity, smaller thermal conductivity or more sensitive interfacial tension variation with temperature is narrower and the local fluid recirculation is stronger than that for the other droplet, which shows a larger migration velocity for the former droplet. These results are consistent with the prediction of [1].

The situation of two spheres with unequal radii is illustrated in figure 6, which corresponds to the case of $a_2/a_1 = 2.0$ and $\lambda = 0.5$. Again, as expected from [1], the larger droplet migrates faster. It is noted that the local recirculation in the vicinity of the larger droplet is substantial, whereas it might disappear in the vicinity of the smaller droplet. As the radius ratio becomes large, the velocity field is dominated by the large droplet, with the smaller droplet introducing only local perturbations.

Mobility coefficients

The numerical results for the mobility coefficients M_{11} and M_{12} , defined by [19a,b], for the case of two droplets of the *same* fluid with various values of η^* , k^* , a_2/a_1 and λ are presented in tables 3-6. Note that M_{21} and M_{22} for the case of $a_2/a_1 = 0.5$ are equal to M_{12} and M_{11} for the case of $a_2/a_1 = 2.0$, respectively. If both droplets are equal in radius, they will move at the same velocity because $M_{21} = M_{12}$, $M_{22} = M_{11}$ and $U_2^{(0)} = U_1^{(0)}$. As expected, the results illustrate that the droplets' interaction decreases rapidly, for all values of η^* , k^* and a_2/a_1 , with an increase in the gap between them (i.e. decreasing λ). However, the interaction between droplets can be very strong when the surface-to-surface spacing gets close to zero. The effect of the interaction, in general, is far greater on the smaller of the two droplets than on the larger one (M_{12} increases dramatically with increasing a_2/a_1) for any given values of η^* , k^* and λ . The larger droplet is hardly influenced by the presence of the smaller one for cases of radius ratio >2.0, unless they are very close together.

Using a method of reflections, Anderson (1985) obtained the mobility coefficients for the coupled thermocapillary motion of two droplets of the same fluid oriented arbitrarily with the temperature gradient. For the axisymmetric motion considered here, his results give

$$M_{11} = 1 - 2\left(\frac{1-k^*}{2+k^*}\right)\left(\frac{a_2}{r_{12}}\right)^3 + \left[4\left(\frac{1-k^*}{2+k^*}\right)^2 - \frac{3}{2}\left(\frac{2+5\eta^*}{1+\eta^*}\right)\right]\frac{a_1^3a_2^3}{r_{12}^6} + O(r_{12}^{-8})$$
[20a]

and

$$M_{12} = \left(\frac{a_2}{r_{12}}\right)^3 - \left[2\left(\frac{1-k^*}{2+k^*}\right) - \frac{9}{2}\left(\frac{1-k^*}{3+2k^*}\right)\left(\frac{2+3\eta^*}{1+\eta^*}\right)\right]\frac{a_1^3a_2^3}{r_{12}^6} + O(r_{12}^{-8}).$$
 [20b]

The mobility coefficients M_{11} and M_{12} calculated from the above asymptotic solution, with the $O(r_{12}^{-8})$ term neglected, are also listed in tables 3-5 for comparison. It can be found that the asymptotic formulas [20a,b] from the method of reflections agree very well with the exact results as long as the droplet surfaces are more than 2/3 of the sum of radii apart (i.e. $\lambda \leq 0.6$). However, accuracy begins to deteriorate, as expected, when the droplets are closed together (say, $\lambda \geq 0.8$). Formula [20b] always underestimates the droplets' interaction.

Note that, as illustrated in table 3, both the exact and the asymptotic solutions predict the following relation for the case of two gas bubbles ($\eta^* = 0$, $k^* = 0$) with arbitrary radii:

$$M_{11} + M_{12} = 1. [21]$$

For the thermocapillary motion of two bubbles of different sizes, the one with the smaller radius (i.e. smaller velocity) is enhanced by the motion of the other, which is retarded at the same time by the motion of the former one. In view of this, it is not surprising that if the bubbles are of equal size (so $U_1^{(0)} = U_2^{(0)}$), both will move with exactly the velocity that would exist in the absence of one

Table 3. T thermocapi asyr	he mobilit llary motic nptotic solu	y coefficients in of two gas ition are evalu	M_{11} and M_{12} s bubbles (η^*) nated from [20]	(defined by $= 0, k^* = 0$); $= 0, b^* = 0$;	[19a]) for the the results of arison	Table 4. T	he mobility of two	coefficients <i>h</i> droplets of id	H_{11} and M_{12} for entical fluid (η	r the thermoca $1^* = 1, k^* = 1$	pillary motí
		Exact so	olution	Asymptoti	c solution			Exact s	solution	Asymptoti	ic solution
a_2/a_1	۲	M ₁₁	M ₁₂	M	M_{12}	a_2/a_1	r	M_{11}	M ₁₂	M	M_{12}
0.5	0.2	0.999702	0.000298	0.999702	0.000298	0.5	0.2	966666.0	0.000296	9666660	0.000296
	0.4	0.997494	0.002506	0.997540	0.002460		0.4	0.999748	0.002373	0.999764	0.002370
	0.6	0.990455	0.009545	0.990976	0.009024		0.6 2	0.997342	0.008122	0.997312	0.008000
	0.8	0.971835	0.028165	0.975284	0.024716		0.8 0.8	0.984568	0.021701	0.984897	0.018963
	0.9	0.951791	0.048209	0.961336	0.038664		0.9	0.964913	0.039570	0.969382	0.027000
	0.95	0.933913	0.066087	0.952112	0.047888		ce.0	0.942951	0.062102	0.95/649	0.031/150 2092200
	0.07	6/ 5776.0	0.07421	0045600	0.054201		0.9/ 0.08	0.926905	0/ 002200	0102620	0.032859
	0.00	0.914001	700900 0	0.943400	0.056600		0.99	0.896151	0.119927	0.945758	0.035937
	0.995	0.893631	0.106369	0.942218	0.057782		0.995	0.880487	0.141515	0.944094	0.036484
10	0.2	0.998996	0.001004	866866.0	0.001002	1.0	0.2	0.999995	0.001000	0.999995	0.001000
	0.4	0.991793	0.008206	0.991872	0.008128		0.4	0.999642	0.008011	0.999664	0.008000
	0.6	0.970383	0.029616	0.971542	0.028458		0.6	0.995552	0.027505	0.996173	0.027000
	0.8	0.917552	0.082447	0.927808	0.072192		0.8	0.969778	0.073473	0.978496	0.064000
	0.9	0.861745	0.138255	0.892268	0.107732		0.9	0.922597	0.129161	0.956405	0.091125
	0.95	0.812366	0.187634	0.869856	0.130144		0.95	0.863894	0.190927	0.939700	0.107172
	0.97	0.781446	0.218554	0.859886	0.140114		0.97	0.818174	0.237044	0.931670	0.114084
	0.98	0.760073	0.239927	0.854668	0.145332		0.98	0.783735	0.272345	0.927333	0.117649
	0.99	0.729203	0.270796	0.849291	0.150709		0.09 0 00 0	0.728810	0.327567	0.922769 0.920309	0.121287
	CKK.N	0.104222	011667.0	110040.0				170700.0		((()))	
2.0	0.2	0.997627	0.002372	0.997628	0.002372	2.0	0.2	0.999996	0.002370	0.999996	0.002370
	0.4	0.980878	0.019121	0.980947	0.019053		0.4	0.999726	0.018981	0.999764	0.018963
	0.6 2.5	0.933692	0.066307	0.934976	0.065024		0.0	0.996146	0.064802	215/00.0	0.064000
	8.0	0.828440	0.1/1004	0.842343	104/01.0		0.0	0.90/649	0.1001/0	0.96469/	0.151.0
	0.9	0.121192	0.257085	0557//0	0.270171		0.05	0.814675	01771770	0.057640	0.254037
	CC-0	0212020	0.407829	0.711296	0 288704		0.97	0.740148	0.444256	0.952010	0.270422
	0.98	0.557914	0.442086	0.701686	0.298314		0.98	0.680392	0.495084	0.948964	0.278872
	0.99	0.509545	0.490455	0.691840	0.308160		0.99	0.583586	0.571350	0.945758	0.287496
	0.995	0.471308	0.528692	0.686828	0.313172		0.995	0.499117	0.633810	0.944094	0.291874
5.0	0.2	0.995370	0.004630	0.995370	0.004630	5.0	0.2	0.999999	0.004630	666666.0	0.004630
	0.4	0.962920	0.037080	0.962941	0.037059		0.4	0.999926	0.037045	0.999942	0.037037
	0.6	0.874242	0.125758	0.874750	0.125250		0.6	0.998753	0.125406	0.999344	0.125000
	0.8	0.695029	0.304971	0.702299	0.297701		0.8	0.985260	0.304457	0.996313	0.296296
	0.9	0.547675	0.452325	0.575277	0.424723		0.9	0.941096	0.457182	0.992525	0.421876
	0.95	0.440799	0.559201	0.499894	0.500106		0.95	0.858238	0.576745	0.989660	0.496167
	0.97	0.382138	0.617862	0.467367	0.532633		0.97	0.777067	0.648116	0.988284	0.528170
	0.98	0.344653	0.65554/	8/ COCF 0	0.549422		0.98	050CU/.U	40006070	0401240	0/00000
	0.99	0.294716	0./05284	0.455451	4000000		0.00 0	104080.0	0.194872	101000.0	+7010C.U
	CKY.0	4200C7'N	U. /40011	0.1424.0	C07C/C.0		<i>LTT.</i> U	0.402010	0.4010.0	UCCU07.U	0.01000

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thermocap	illary motio	n and the moti fluid $(\eta$	on under gravit $* = 10, k^* = 1$)	y of two dropl	ets of identical
		Exact s	olution	Exact se	olution
		(thermocapil	lary motion)	(motion und	ier gravity)
a_2/a_1	۲	M ₁₁	M_{12}	M ₁₁	M_{12}
0.5	0.2	0.999995	0.000296	0.999873	0.094246
	0.4	0.999696	0.002374	0.998156	0.184820
	0.6	0.996660	0.008202	0.992107	0.269224
	0.8	0.981258	0.023170	0.979940	0.350663
	0.9	0.957534	0.045287	0.970164	0.396523
	0.95	0.930994	0.073931	0.962657	0.425311
	0.97	0.912272	0.096542	0.958391	0.439711
	0.98	0.898942	0.113736	0.955701	0.448160
	0.99	0.880316	0.139192	0.952325	0.458109
	0.995	0.866995	0.158399	0.950147	0.464130
10	0.2	0.999993	0.001000	0.999674	0.141463
	0.4	0.999531	0.008019	0.994990	0.278040
	0.6	0.994237	0.027842	0.976310	0.407312
	0.8	0.960831	0.079053	0.929174	0.538609
	0.9	0.897597	0.149654	0.882788	0.618792
	0.95	0.817749	0.232187	0.844579	0.672238
	0.97	0.758256	0.292418	0.822806	0.699824
	0.98	0.714937	0.335986	0.809254	0.716223
	0.99	0.653569	0.397482	0.792598	0.735686
	0.995	0.609330	0.441771	0.782152	0.747521
2.0	0.2	266666.0	0.002370	0.999477	0.188491
	0.4	0.999638	0.018993	0.991587	0.369641
	0.6	0.994899	0.065344	0.956830	0.538447
	0.8	0.956897	0.174866	0.855715	0.701327
	0.9	0.868816	0.303115	0.747139	0.793046
	0.95	0.742083	0.435043	0.656115	0.850621
	0.97	0.640184	0.524592	0.604564	0.8/9423
	0.98	0.562755	0.587009	0.572758	0.896321
	0.99	0.448818	0.672518	0.554084	0.910219
	0.995	0.363949	0.732420	0.510153	0.928260
5.0	0.2	666666.0	0.004630	0.999483	0.235143
	0.4	1066660	0.037052	0.991343	0.458119
	0.6	0.998320	0.125679	0.951918	0.657509
	0.8	0.979802	0.309343	0.817568	0.826102
	0.9	0.917920	0.475614	0.653309	0.901203
	0.95	0.800756	0.614731	0.506318	0.940161
	0.97	0.688026	096669.0	0.420618	0.957786
	0.98	0.592460	0.757110	0.367065	0.967740
	0.99	0.436879	0.833683	0.301398	0.979261
	0.995	0.309079	0.886971	0.260614	CU2086.U

Table 5. 7	The mobility of two	/ coefficients <i>h</i> droplets of ide	I_{11} and M_{12} for entical fluid (η^{4}	the thermoca $= 1, k^* = 10$	pillary motion
		Exact s	olution	Asymptoti	c solution
a_2/a_1	۲	M.1	M ₁₂	M11	M ₁₂
0.5	0.2	1.000442	0.000294	1.000442	0.000294
	0.4	1.003428	0.002248	1.003421	0.002240
	0.6	1.010681	0.006764	1.010464	0.006514
	0.8	1.022149	0.014191	1.019814	0.010614
	0.9	1.027988	0.023367	1.023004	0.010074
	0.95	1.027417	0.038644	1.023431	0.008343
	0.97	1.023170	0.054865	1.023281	0.007274
	0.98	1.017466	0.070629	1.023125	0.006646
	0.99	0.995962	0.100281	1.022910	0.005952
	0.995	0.988835	0.137303	1.022780	0.005580
1.0	0.2	1.001497	0.000977	1.001497	0.00097
	0.4	1.011807	0.007820	1.011808	0.007814
	0.6	1.038377	0.025209	1.038313	0.024884
	0.8	1.084713	0.058241	1.083712	0.052113
	0.9	1.110690	0.091742	1.111776	0.067026
	0.95	1.111051	0.131367	1.126300	0.073838
	0.97	1.095715	0.166627	1.132081	0.076312
	0.98	1.075446	0.198372	1.134950	0.077479
	0.99	1.028172	0.258767	1.137799	0.078595
	0.995	0.971406	0.323070	1.139215	0.079131
2.0	0.2	1.003553	0.002368	1.003553	0.002368
	0.4	1.028301	0.018836	1.028310	0.018832
	0.6	1.094324	0.062832	1.094460	0.062514
	0.8	1.217123	0.150397	1.218925	0.143355
	0.9	1.293309	0.228540	1.306504	0.199075
	0.95	1.307492	0.298971	1.356855	0.230626
	0.97	1.280751	0.349891	1.378210	0.243893
	0.98	1.240522	0.391005	1.389144	0.250659
	0.99	1.142310	0.463206	1.400249	0.257511
	0.995	1.021665	0.536076	1.405865	0.260969
5.0	0.2	1.006944	0.004629	1.006944	0.004629
	0.4	1.055519	0.037006	1.055523	0.037005
	0.6	1.187025	0.124788	1.187125	0.124637
	0.8	1.439801	0.298036	1.442338	0.294258
	0.9	1.609693	0.435258	1.628542	0.417743
	0.95	1.669512	0.533021	1.738343	0.490451
	0.97	1.649227	0.588461	1.785560	0.521693
	0.98	1.600063	0.626469	1.809893	0.537788
	0.99	1.462791	0.684165	1.834719	0.554204
	CKY.U	C4/082.1	0.736467	1.847328	0.562540

of the droplets for any value of λ (Meyyappan *et al.* 1983; Anderson 1985). It should be noted that, while there is no net interaction between the bubbles, there are certainly perturbations to the flow field due to their proximity.

For the specific case of two identical liquid droplets with equal radii tables 4-6 give the relation $M_{11} + M_{12} > 1$, which indicates that each droplet moves faster than its undisturbed velocity $U^{(0)}$. This enhancement in droplet velocity is similar to the observation when the driving force for droplet motion is gravity (Rushton & Davies 1973, 1978). When the ratio of thermal conductivity between the internal and surrounding fluid, k^* , becomes larger, M_{12} decreases and $M_{11} + M_{12}$ increases. On the contrary, M_{12} increases and $M_{11} + M_{12}$ decreases if η^* is increased. Note that, when the two liquid droplets differ in size, the one with the larger radius (i.e. larger velocity) can be slowed down to a speed less than its undisturbed value.

Numerical results of M_{11} , M_{12} , M_{21} and M_{22} for a typical case of two droplets of different fluids are given in table 7: again, the droplets' interaction is stronger when they are closer together. The effect of interaction is also greater on the small droplet than on the larger one. In general, the droplet with the smaller velocity is speeded up by the motion of the other droplet.

For the motion of two spherical droplets on which a body force (e.g. a gravitational field) is imposed along their line of centers, the exact results of the droplet velocities were developed using bipolar coordinates by Rushton & Davies (1973, 1978). Although their theoretical analysis is

_		01 991	-, 12 , 2	,	
a_2/a_1	î.	M ₁₁	<i>M</i> ₁₂	M ₂₁	M ₂₂
0.5	0.2	1.000441	0.000296	0.002368	0.999996
	0.4	1.003322	0.002373	0.018807	0.999726
	0.6	1.009394	0.008122	0.062602	0.996146
	0.8	1.013750	0.021700	0.151325	0.967849
	0.9	1.007501	0.039571	0.238062	0.904504
	0.95	0.993362	0.062105	0.325634	0.814625
	0.97	0.979825	0.080576	0.391364	0.740148
	0.98	0.968168	0.095494	0.443134	0.680392
	0.99	0.948075	0.119927	0.527346	0.583586
	0.995	0.929514	0.141515	0.602040	0.499117
1.0	0.2	1.001495	0.000250	0.001000	0.999995
	0.4	1.011647	0.008011	0.007779	0.999642
	0.6	1.036258	0.027505	0.024815	0.995552
	0.8	1.068409	0.073473	0.057857	0.969778
	0.9	1.065309	0.129161	0.097629	0.922597
	0.95	1.029117	0.190927	0.151366	0.863894
	0.97	0.988153	0.237044	0.198864	0.818674
	0.98	0.951028	0.272345	0.239094	0.783735
	0.99	0.885213	0.327567	0.307878	0.728810
	0.995	0.823475	0.374473	0.371044	0.682027
2.0	0.2	1.003552	0.002370	0.000294	0.999996
	0.4	1.028179	0.018980	0.002191	0.999765
	0.6	1.092469	0.064802	0.006473	0.997347
	0.8	1.198824	0.166196	0.013329	0.984597
	0.9	1.231839	0.272266	0.024742	0.964913
	0.95	1.182584	0.374498	0.046660	0.942951
	0.97	1.107672	0.444256	0.069164	0.926903
	0.98	1.034573	0.495084	0.089457	0.914810
	0.99	0.899464	0.571349	0.125712	0.896151
	0.995	0.769470	0.633810	0.160078	0.880487
5.0	0.2	1.006943	0.004630	0.000036	0.999999
	0.4	1.055485	0.037046	0.000261	0.999946
	0.6	1.186399	0.125406	0.000651	0.999430
	0.8	1.430383	0.304457	0.000783	0.997052
	0.9	1.564555	0.455417	0.002112	0.992157
	0.95	1.551447	0.576773	0.005764	0.991492
	0.97	1,462447	0.648096	0.010413	0.989506
	0.98	1.359027	0.696457	0.015082	0.988000
	0.99	1.147966	0.764971	0.024203	0.985692
	0.995	0.930806	0.818474	0.033520	0.983786

Table 7. The exact solution of the mobility coefficients M_{11} , M_{12} , M_{21} and M_{22} (defined by [19a,b]) for the thermocapillary motion of two droplets $(n^* = 1, k^* = 1, n^* = 1, k^* = 10)$

legitimate, some typographical errors occur in their formulas for droplet interactions. The correct mobility coefficients M_{11} and M_{12} for droplet movement in a gravitational field are also presented in table 6 for comparison. Clearly, the interaction effect in thermocapillary motion is much weaker (smaller in M_{12}) than for motion under gravity. This result is consistent with the predictions by Meyyappan & Subramanian (1984) and Anderson (1985).

CONCLUSIONS

The thermocapillary motion of two fluid droplets along their line of centers has been examined in this work. The temperature and velocity fields are solved using bipolar coordinates and the droplet velocities are obtained for various values of the fluid properties, droplet sizes and separation distance. It is found that the interaction between droplets can be very strong when their gap thickness approaches zero. The asymptotic formula [20b] generated from the method of reflections always gives too small an effect of droplet interactions, and the error can be significant when the droplets are near contact. The influence of the interaction is far greater on the smaller of the two droplets. The droplet with the smaller velocity is enhanced by the motion of the other, which can be retarded simultaneously by the motion of the first droplet. For the special case of two identical liquid droplets, both migrate with the same velocity, which is larger in magnitude than that which would exist in the absence of one of the droplets. There is no interaction between two bubbles of equal size. In general, the effect of droplet interactions on thermocapillary motion is much weaker than that on the motion in a gravitational field.

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