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Drop motions and interfacial instability

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Abstract. Drops and bubbles are ubiquitous. Stokes, Rybczynskii, Hadamard, Boussinesq and others provided the drag law experienced by a drop. Young, Goldstein and Block and others studied the motion due to interfacial stresses induced on a drop immersed in a medium where an imposed thermal gradient exists. They demonstrated the possibility of its levitation in the presence of gravity. Levich, Sanfeld and others pointed out the role played by surfactants in affecting the drag law and the possible fluid motion inside the drop. All those works refer to *passive* drops, i.e. drops experiencing at most the interfacial stresses due to variation of interfacial tension with temperature or surfactant concentration in the surrounding fluid near the drop surface. After providing a succinct account of the results of the earlier theories and some relevant experiments, we consider the behaviour of an *active* drop, i.e. a drop with chemical reaction at its surface or with an internal heat generation source, etc. Attention is focused on the case of a drop immersed in a *homogeneous* surrounding when due to surface stresses (the Marangoni effect) and, consequently, due to thermo/soluto-hydrodynamic instability there is spontaneous breaking of the radial symmetry of the temperature and/or concentration distributions, hence overcoming the drag and originating self-sustained translational drop motion. Moreover, the autonomous motion may offer a multiplicity of steady values for a given external (weak) force like buoyancy, and levitation is possible for multiple (weak) buoyancy levels.

1. Introduction; the role of surface tension and surface tension gradients

When there is an open surface or an interface exists between two liquids, the surface or interfacial tension accounts for the jump in normal stresses proportional to the surface curvature across the interface, and hence affects its shape and stability. Gravity competes with the Laplace forces in accommodating equipotential levels with curvature. Their balance permits, for instance, the stable equilibrium of the spherical shape of drops and bubbles.

When surface tension varies with temperature or composition and, consequently, with position along an interface, its change takes care of the jump in the tangential stresses. Hence its gradient acts like a shear stress applied by the interface to the adjoining bulk liquid (Marangoni stress), and thereby generates flow or alters an existing one. Surface tension gradient-driven flows are known to also affect the evolution of growing fronts, and measurements of transport phenomena. The variation of surface tension along an interface may be due to the existence of a thermal gradient along the interface or perpendicular to it. In the former case, we have instantaneous convection, while in the latter, flow occurs past an instability threshold.

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To qualitatively assess the role of gravity relative to forces induced by surface tension or its gradient, some dimensionless groups help. There is the (static) Bond number $Bo = gl^2\,\Delta\rho/\sigma$, where $\Delta\rho$ is the density difference between two fluids (think of a drop of a fluid immersed in another liquid), σ is the interfacial tension and l is the space scale involved in the problem. In Earth-based experiments, gravity overcomes surface tension effects when l and $\Delta\rho$ are large enough. With surface tension gradients we have the modified (dynamic) Bond number $Bo^* = gl^2\,\Delta\rho/\Delta\sigma$, although thermal gradients are generally accounted for by the Marangoni number, $Ma = l\,\Delta\sigma/\eta\chi = Pr\,Re_\sigma$, where η is the dynamic viscosity ($\eta = \rho\nu$), χ is thermal diffusivity, $Pr = \nu/\chi$ is the Prandtl number, and Re_σ denotes a surface Reynolds number, with the gradient-induced velocity scale $V = \Delta\sigma/\eta$, and $\Delta\sigma = (\mathrm{d}\sigma/\mathrm{d}T)\Delta T$. Thus the Marangoni number is a Peclet number, $Pe = Vl/\chi$, in the heat equation—this is quite like how the standard Reynolds number appears in the Navier–Stokes (momentum) equations. Other forms of Peclet number may be of use according to the velocity scale involved in the problem.

Surface tension gradient-driven flows are, generally, laminar flows, with or without cellular patterns, like in Bénard convection [1–3]. For interface and bulk fluid moving tied together, only high enough Marangoni numbers lead to turbulent flows with high dissipation. As for flows driven by capillary forces the Peclet/Marangoni number appears in the heat equation; then for high Prandtl number fluids the velocity field is slaved by the temperature while for low Prandtl number fluids the latter slaves the former. In flows with very low Re, inertia can be neglected in the flow (e.g. Stokes creeping flow) of very viscous fluids like the silicone oils, where however possible high values of Ma exist. Consequently, Reynolds (Kolmogorov) turbulence, which is mostly inertial, appears as a different regime from the interfacial turbulence or space-time chaos in capillary-driven flows (with high Pr) where there is strong dissipation—hence the interest in proceeding to high-Peclet/Marangoni-number flows to explore new features of turbulence with the numerical approach and benefiting from the rather recent opportunity offered by low-g experimentation.

High-Peclet/Marangoni-number flows, and thus apparently predominant surface tension gradient-driven flows in Earth-based experiments, are different from analogous low-g flows. For instance, in an externally imposed thermal gradient, for the instantaneous surface tension gradient-driven flow of a drop or a bubble to be steady in the presence of gravity, a non-zero hydrodynamic force is needed, which is quite a different case from that for low-g conditions. Hence Earth-based and low-g Marangoni flows of drops or bubbles are different. Indeed, gravity-induced motions alter the temperature field in the fluid surrounding the drop or bubble, thus changing the Marangoni forces acting on it.

Since the generation of a surface tension inhomogeneity is related to the imposition of thermal gradients, in Earth-based experiments buoyancy-induced flows can also occur simultaneously with surface tension gradient-driven flows. Then the relative magnitude of surface tension gradient and buoyancy forces is given by the inverse of Bo^*. Surface deformation and surface curvature appear in the complete balances of tangential and normal stresses, respectively. The former balance yields a capillary number $Ca = \eta V/\sigma$, that, using V as earlier defined, accounts for the relevance of surface deformation effects in a surface tension gradient-driven flow. Instantaneous motion caused by purely capillary forces provokes the surrounding fluid to move towards the cooler region while the drop or bubble moves towards the warmer one. When surface tension is not constant a spherical drop or bubble tends to become deformed due to the normal-stress (im)balance at the interface. At least in capillary-driven creeping flow, the curvature change is of order $\Delta\sigma/\sigma$, i.e. of order Bo/Bo^*, which is the capillary number, and as Ca is much smaller than unity, deformation is not really relevant.

The surface tension variation acts dramatically on interphase transport processes, evaporation, adsorption/desorption processes, drop and bubble migration, etc. Indeed, when a highly surface-active substance is strongly adsorbed at an interface the resulting surface film tends to alter transport rates either hydrodynamically, by locally damping or enhancing eddies and ripples, or by causing stagnation over a considerable portion of the interface, or if the molecules form a highly packed film by imposing an (energy) barrier to the passage of other molecules across the interface. Even minute traces of surfactant are known to produce dramatic effects at both quantitative and qualitative levels. Theory and Earth-based experiments indicate that transport processes, surface chemical reaction processes, catalysis phenomena, ... can be altered by either buoyancy-driven or surface tension gradient-driven flows, or both appearing together, due to even mere ppm changes in composition. For instance, when a surfactant changes the surface tension, interfacial instability can appear during transfer out of the phase of higher kinematic viscosity or lower solute diffusivity. Amplification of interfacial convective motions can also be promoted by large $\Delta\sigma$-changes with composition, large thermal gradients, and large differences of kinematic viscosity and diffusivity between phases.

In this paper we will concentrate on the specific dynamics of (passive or active) drops or bubbles immersed in another fluid. Drops and bubbles are ubiquitous. Liquids used to power space vehicles or used in Shuttle cooling systems, upon evaporation lead to bubbles—hence the need of capillary forces, and eventually surface tension gradient-driven flows, to manage bubbles, isolated or in clusters following aggregation and possible coalescence for low g. When one phase is dispersed in another as a drop or bubbles, the approach of an eddy causing a surface deformation and a local reduction in surface tension can cause violent motion which in turn creates other eddies. The coalescence of drops with either different solutes or different solute concentrations, or the introduction of jets of solvent, can cause similar sudden movements, altering mass transfer. Boiling leads to formation of relatively small bubbles that in Earth-based experiments float, and move up but distribute all over the container, while in low-g conditions bubbles tend to coalesce, thus forming bigger ones that drastically affect the overall process. Clearly, drop dynamics is not just of mere fluid physics interest. It is of relevance in atmospheric sciences, chemical engineering, materials science, etc. Understanding some aspects of drop dynamics has led to the most spectacular albeit simple technological spin-off of microgravity space research.

2. Drops and bubbles; hydrodynamic drag

A liquid drop is a model for many natural systems as disparate as planets, nuclei, etc. As indicated by the definition of the Bond numbers given earlier, it is also a g-detector. Drop dynamics provides the test ground for theoretical analyses (e.g. using matched asymptotic expansions), experimental techniques and numerical methods (which on occasion perform better than real experiments). The study of drops also allows one to sequentially proceed from simple to complex.

From Newton's experiments in 1710, theory and later observations, the magnitude of the drag force on solid drops/spheres in steady motion of a viscous fluid was given as

$$F_D = 0.22\pi a^2 \rho U_\infty^2 \tag{1}$$

where U_∞ is the relative velocity of the particle and fluid, a is the particle radius, and ρ is the fluid density. This kinetic theory relation is for 'large' values of U_∞, for which inertial effects are important.

Stokes, in 1850, suggested that at very low relative velocities the inertial effects are so small that they can be omitted from the Navier–Stokes equations. Under these conditions, the total drag on a solid sphere is

$$F_D = 6\pi\eta a U_\infty. \tag{2}$$

In 1911–12 Rybczynskii and Hadamard, independently, solved the Stokes problem for a liquid drop with flows outside and inside. Their extension of (2) is

$$F_D = 4\pi \frac{1 + 3\beta/2}{1 + \beta} \eta_1 a U_\infty \tag{3}$$

with $\beta = \eta_2/\eta_1$, where the subscripts $i = 1, 2$ correspond to the liquids outside and inside the drop, respectively. Clearly, the limit η_2 going to infinity yields back Stokes's law (2), while $\eta_1 \gg \eta_2$ yields the corresponding law for a bubble, with the factor 6 being replaced by 4 in (2) [4].

Oseen pointed out that at a great distance from the sphere the inertial terms become as important as the viscous terms, and suggested a possible improvement of the Stokes law (2) by taking inertial terms partly into consideration [5]. His drag force is

$$F_D = 6\pi\eta a U_\infty \left(1 + \frac{3}{8} Re\right) \tag{4}$$

where $Re = U_\infty a/\nu$. Today we know that neither Stokes's nor Oseen's laws are uniformly valid. Rather Stokes's analysis is valid in a small enough neighbourhood of the sphere (creeping flow at zero Reynolds number) and Oseen's analysis though valid for matching at the flow velocity values far from the sphere is not valid when approaching it. The Oseen approximation although incorporating inertial terms is still a linear theory. Several authors did attempt, not always successfully, an improvement upon the Oseen and Stokes analyses. It was not until 1957 that Stokes's and Oseen's results were properly put in context and generalized, thanks to the works of Proudman and Pearson [6] and Kaplun and Lagerstrom [7], who developed the matched-asymptotic-expansions method for the flow around the sphere. The basic concept was to consider Stokes's solution as a local (inner) solution of the problem and Oseen's as a regular (outer) solution rather than considering Oseen's as an improvement upon Stokes's. The inner solution was assumed to be valid in a spherical region of radius $1/Re$ around the sphere while the outer solution was valid from infinity down to the $1/Re$ neighbourhood. In the overlapping zone both solutions were accepted as valid—hence the need for appropriately matching them. Proudman and Pearson found that for flows with non-vanishing although low Reynolds number ($Re \ll 1$), the hydrodynamic drag on the sphere is

$$F_D = 6\pi\eta a U_\infty \left(1 + \frac{3}{8} Re + \frac{9}{40} Re^2 \ln Re + \mathrm{O}(Re^2)\right) \tag{5}$$

which shows the non-analytic form of expansion. The scheme of Proudman and Pearson was also applied to the problem of heat/mass transfer to (from) a sphere (see e.g. [8, 9]). Acrivos and Taylor [8] considered the convective terms in the heat equation while using the Stokes flow. They obtained a Peclet number ($Pe = U_\infty a/\chi$) expansion of the (convective) Nusselt number:

$$Nu = 1 + \frac{1}{2} Pe + \frac{1}{2} Pe^2 \ln Pe + 0.415 Pe^2 + \frac{1}{4} Pe^3 \ln Pe + \mathrm{O}(Pe^3) \tag{6}$$

which also shows a non-analytic form.

As already noted for flows driven by capillary forces in high-Prandtl-number fluids, what actually matters is the Peclet number, and the Stokes approximation is a quite valid starting point (at $Re = 0$ the velocity field is slaved by the temperature). Subramanian [10] and later on Merritt [11] solved the problem of the drift of a drop in an external temperature gradient using the matched-asymptotic-expansions method but not leading in this case to a logarithmic term.

Taylor and Acrivos also considered the deformation of the sphere (a bubble) and showed that for it to be noticeable the capillary number must be non-negligible [12]. For a slightly deformable bubble they obtained the radius in terms of the angle θ:

$$a(\theta) = a\left[1 - \frac{5}{48}\,Re\,Ca(3\cos^2\theta - 1)\right] \tag{7}$$

for $Re \ll 1$ and $Ca \ll 1$. Note that at $Re = 0$ (i.e. in the creeping-flow approximation) a drop or a bubble remains spherical irrespective of the low or high value of the (constant) surface tension. Equation (7) shows that a drop or a bubble will be deformed from the spherical shape when inertial effects are taken into account. However, deformation may be relevant even when inertial effects are ignored if as earlier noted the surface tension σ is sufficiently non-uniform.

Different authors have also extended the theory to account for various non-Newtonian fluid properties like first and second normal-stress coefficients for viscoelastic fluids or multi-scales (Oldroyd fluids) and power laws.

Young, Goldstein and Block [13] were the first to realize the possibility of levitating a drop or a bubble by means of capillary forces (Marangoni stresses). They showed that a drop or a bubble placed in a temperature gradient tends to move towards the hotter point. This is the motion of the drop relative to the flow induced along its surface by the lowering of surface tension at its leading pole (hotter than the rear pole). The Marangoni stresses not only help in overcoming drag but even lead to positive (bubble) or negative (heavy-drop) buoyancy, and hence levitation for a sufficiently high temperature gradient. Using the Stokes–Rybczynskii–Hadamard approximation they also computed the terminal velocity of a drop or a bubble in the field of gravity ($Re \ll 1$, $Pe \ll 1$; here these numbers are defined using the far-field velocity scale), and experimentally checked the theoretical prediction within reasonable accuracy (within 20%) with an experiment using rising bubbles in a liquid layer heated from below (diameters $2a = 10^{-3}$–2.2×10^{-2} cm; $\mathrm{d}T/\mathrm{d}z = 10$–$90$ K cm^{-1}). Later on Bratukhin, Briskman, Zuev, Pshenichnikov and Rivkind [14] did a similar experiment with rising bubbles in a laterally heated vertical liquid layer. They experimented using neutrally buoyant liquid water at 4 °C. For the Young, Goldstein and Block problem the balance between capillary, buoyancy and drag forces is

$$F_\sigma + F_g + F_D = 0 \tag{8}$$

with the (Marangoni) capillary force

$$F_\sigma = -\frac{4\pi a^2}{(1+\beta)(2+\delta)}\frac{\mathrm{d}\sigma}{\mathrm{d}T}(\nabla T)_\infty \tag{9}$$

and the buoyancy force

$$F_g = \frac{4}{3}\pi a^3 g(\rho_2 - \rho_1). \tag{10}$$

The drag force F_D was defined in (4). $\delta = \lambda_2/\lambda_1$ is the ratio of thermal conductivities (drop to surrounding fluid).

In their experiment, Young *et al* used a liquid bridge. An improvement eliminating possible capillary convection at the open sides was carried out by Hardy [15]. He used a closed cavity with silicone oil and air bubbles ($2a = (5-25) \times 10^{-3}$ cm; $\mathrm{d}T/\mathrm{d}z = 40$–$140$ K cm^{-1}). Hardy noticed the role of contamination at the surface of the bubble earlier noted by Levich [4]. Further improvement came with an experiment by Merritt and Subramanian [16]. Experimentalists started using drops rather than bubbles. Barton and Subramanian [17] used neutrally buoyant drops ($2a = 20$–600 μm, $\mathrm{d}T/\mathrm{d}z = 2.4$ K mm^{-1}). Recent Earth-based and low-g (TEXUS, D2) work by Braun and colleagues [18] on thermocapillary migration of drops provided the most accurate verification of the Young, Goldstein and Block prediction. They used flows with Peclet/Marangoni numbers in the range 10^{-5}–10^{-6}, but with non-standard surface tension gradient behaviour, i.e. the surface tension increasing with the increase of temperature (2-butoxyethanol–water mixture with liquid/liquid phase separation at 61.14 °C on the lower branch of the closed miscibility gap; $2a = 11$ mm, $\mathrm{d}T/\mathrm{d}z = 36.9$ K m^{-1}, $\mathrm{d}\sigma/\mathrm{d}T > 0$). For an extensive review see [19, 20].

Worth mentioning also are experiments carried out by Neuhaus and Feuerbacher [21] who found disagreement with the Young, Goldstein and Block prediction but who proposed that the solution of the difficulty lay in augmenting the theory with a surface dilatational viscosity, in agreement with a conjecture put forward long ago by Boussinesq (see reference [4]).

The above-given evidence shows how the slightest thermal gradient does induce, via the Marangoni effect, drop motions outside and inside. The case is very much like that of the flow induced by the slightest pressure gradient in the bulk of a liquid.

3. Active drops

Recently, we have taken a new approach to the study of drop (or bubble) motions, linking the classical ideas to the more recent studies of hydrodynamic instabilities, and hence leading to new, fascinating theoretical results. We have studied the influence of the *radial* temperature and/or concentration profiles, which would exist inside and/or outside a motionless drop, on the possible drop motion, when the medium far away from the drop is *homogeneous*. The profiles may be due to chemical reactions [22–30], and uniform heat generation inside a drop [31–33] or on the surface [34], which itself may be due to a chemical reaction or radiation absorption, and also temperature relaxation, when the drop and the outer fluid initially have different temperatures [35]. Losing their spherical symmetry due to motion of fluids inside and outside the drop, the temperature and/or concentration distributions promote the appearance of the Marangoni stresses on the drop surface, hence dramatically affecting the drop behaviour. For instance, we have shown that due to the radial profiles the motionless state of fluids inside and outside the drop in the absence of buoyancy can be unstable to convection, and a countable number of linear unstable modes appears, each one having its own steady instability threshold [25, 28, 33]. The fundamental mode includes drop translation, while the other modes correspond to some flow inside and outside the drop without its translation as a whole. The latter possibility was studied by Sanfeld and collaborators [29, 30]. Up to now several results have been obtained around translational steady instability when the instability thresholds for the other modes are higher than the translational threshold.

For instance, for small enough buoyancy there can be up to three [23–28, 32–34] or up to five [33] stationary regimes of drop motion. The stability analysis shows that only one of the three and two of the five regimes can be stable [25, 33, 36]. In the absence of buoyancy, besides the motionless state, autonomous or self-sustained motion can exist [23–28, 32–34], neutrally stable to velocity perturbations in an orthogonal direction [25, 33, 36]. Another multiplicity can also occur, e.g. the drop can move at a given single velocity under up to three different levels of buoyancy [25, 34]. It has also been shown that levitation of a drop under non-zero buoyancy is possible, when the appropriate balance exists due to the induced flow rather than to some external factor or inhomogeneity [25, 34]. This situation is indeed richer when the self-propulsion exists together with external inhomogeneities [26]. Finally, the interaction of modes yields qualitatively different multiplicities of the motion regimes [37].

4. Interfacial instability

Let us consider, for instance, a drop with uniform internal heat generation suspended in an infinitely extended homogeneous liquid. If the liquids inside and outside the drop, as well as the drop as a whole, are at rest, a radial temperature distribution is established, with the hottest temperature in the centre of the drop and the coolest temperature far away from the drop. As soon as due to some spontaneous fluctuation or the action of a bias field like buoyancy the drop is set in motion, the radial symmetry of the temperature profile breaks down, thus inducing a non-homogeneous temperature distribution along the drop surface, which in turn couples to the initial fluctuation that may be reinforced. Indeed, the tangential (Marangoni) stresses produced by the surface tension variation with temperature should be compensated by an additional motion of the liquids; hence, the Marangoni effect plays the role of feedback mechanism.

Now let us observe what effect on the motion of the drop could be produced as a whole. This largely depends on the temperature difference that arises between its leading and trailing poles. As the surface tension usually decreases with increasing temperature, when the leading temperature is lower than the trailing one, the Marangoni stresses act from the trailing pole to the leading one, thus increasing the resistance to the motion, a phenomenon discussed by Levich [4] in the context of surface contamination by surfactants. In the opposite case, when the temperature at the leading pole is higher than that at the trailing pole, the Marangoni stresses act, helping the drop to keep moving. If the temperature difference is strong enough to overcome dissipative effects (a ratio that is measured in terms of the Marangoni number), the drag force usually accompanying the motion of objects in a liquid medium is replaced for our drop by thrust. Thus various regimes may be found when the drop moves *against* the mass forces. Under free-fall conditions, the motionless state of the drop can become unstable and the drop spontaneously starts to move and keeps steadily moving—hence an *autonomous* or *self-sustained* motion ensues. In the absence of buoyancy or any other external force all directions in space are allowed for this autonomous motion. Then the breakdown of this symmetry can only be induced by the initial conditions. When a force is present, and if it is weak enough, a multiplicity of steady regimes of the drop motion persist. Actually, one of the regimes is the low-velocity unstable regime, which proceeds from the motionless state, slightly shifted by the force. Two other possible regimes are with velocities close to the autonomous motion velocity. On the other hand, if the force is strong enough, the multiplicity associated with the autonomous motion is suppressed and there remains only a single motion regime.

Coming back to the drop with uniform internal heat generation, let us identify the factors determining the sign of the temperature difference. There is competition between two mechanisms. As the temperature at infinity is lower than that at the surface of the drop, the flow brings cooler fluid to its leading pole. However, as the temperature inside the drop is higher than that at the surface, internal circulation in the drop brings fluid at higher temperature to the leading pole. The result of this competition depends on the ratio of the thermal diffusivities inside and outside the drop. When the latter is high enough the convective heat transport outside is not important as compared to that inside, and the leading pole becomes hotter than the trailing one. If in addition the heat generation is strong enough, and hence the Marangoni number is high enough, then the absolute value of the temperature difference helps in overcoming the dissipation, thus promoting the effects discussed earlier [33]. An order-of-magnitude estimate yields that for a drop of 1 mm diameter, several degrees kelvin in temperature difference suffice for reaching the instability threshold.

Until now we have only discussed the influence of the Marangoni effect on the motion of the drop as a whole. However, the motionless state of liquids can also be unstable to convection inside and outside the drop without global translation of the drop. The mechanism is again that the flow causes a redistribution of temperature at the surface and hence promotes the motion. There exist a countable set of modes characterized by the number of circulation zones inside the drop. Each mode has its own steady instability threshold that can be expressed in terms of a critical Marangoni number. For the higher-order modes, when the number of circulation zones is sufficiently large, the analogy with the problem of Bénard–Marangoni instability in a system of two horizontal infinitely deep liquid layers subjected to a vertical temperature gradient [38] becomes apparent. Indeed, let us focus on the ratio of thermal diffusivities. For the two-layer system we know that the steady instability occurs only if the thermal diffusivity of the hotter phase is just barely smaller than that of the colder phase. The same argument holds for the high-order modes in the drop. However, for low-order modes this condition should be modified. In the latter case we can only demand that the thermal diffusivity of the hotter phase is small *enough*. A more precise answer demands some calculations. The reason for this is the geometrical asymmetry of the phases. Just by analogy with the two-layer case where oscillatory Marangoni instability was predicted [38], an oscillatory instability should be also expected for the drop motion.

5. Linear modes and their steady Marangoni instability thresholds

The stationary motion of a drop in a large extended fluid is considered. We assume that both the Reynolds and the Peclet numbers are small and that the drop is spherical. The fluids inside and outside the drop are treated as Newtonian and incompressible. A reference frame where the drop is motionless is used. In the leading order the flow field inside and outside the drop obeys the Stokes equation

$$E^2 E^2 \psi_i = 0 \tag{11}$$

with

$$E^2 = \frac{\partial^2}{\partial r^2} + \frac{\sin^2\theta}{r^2}\frac{\partial^2}{\partial(\cos\theta)^2}. \tag{12}$$

Here ψ is a stream function defined as

$$v_r = \frac{1}{r^2\sin\theta}\frac{\partial\psi}{\partial\theta} \qquad v_\theta = -\frac{1}{r\sin\theta}\frac{\partial\psi}{\partial r}$$

and r, θ are the radial and angular spherical coordinates (the origin is at the centre of the drop), and v_r and v_θ are the radial and angular components of the velocity field. As earlier, the subscripts $i = 1, 2$ correspond to the regions inside and outside the drop, respectively.

Far away from the drop we have a uniform flow with a velocity U_∞ just opposite to that of the drop in the laboratory frame of reference, i.e.

$$\psi_1 \to U_\infty r^2 \sin^2\theta/2 \qquad \text{as } r \to \infty. \tag{13}$$

At the drop surface ($r = 1$, where the radial coordinate is non-dimensionalized with the drop radius a) the stick and no-penetration conditions must be satisfied:

$$\psi_1 = \psi_2 = 0 \qquad \partial\psi_1/\partial r = \partial\psi_2/\partial r. \tag{14}$$

Using the general solution of the Stokes equations [39], the solution of our problem (11)–(14) is

$$\psi_{10} = U_\infty\,(r^2 - 1/r)\,G_2(\mu) + \sum_{n=2}^{\infty} A_n (r^{-n+3} - r^{-n+1})\,G_n(\mu) \tag{15a}$$

$$\psi_{20} = \frac{3}{2}\,U_\infty\,(r^4 - r^2)\,G_2(\mu) + \sum_{n=2}^{\infty} A_n\,(r^{n+2} - r^n)\,G_n(\mu) \tag{15b}$$

$$G_2(\mu) = \frac{1-\mu^2}{2} \qquad G_3(\mu) = \frac{\mu(1-\mu^2)}{2} \qquad \cdots$$

where $G_n(\mu)$ $(n = 2, 3, \ldots)$ are the Gegenbauer polynomials of the first kind, of order n and of degree $-1/2$, and $\mu = \cos\theta$. As r is dimensionless, the stream function has the dimension of velocity.

As shown by Happel and Brenner [39], the hydrodynamical force is determined by a single coefficient in the series (15a), (15b):

$$F_{hd} = -4\pi\eta_1 a A_2. \tag{16}$$

The values U_∞ and F_{hd} occurring in (15), (16) are considered positive when they align with the positive direction of the symmetry axis ($\theta = 0$) and otherwise negative. For a drop to be in steady motion the sum of the hydrodynamical and buoyancy forces must vanish. Hence the coefficient A_2 is determined by buoyancy.

To find the unknown coefficients in (15) we use the tangential-stress balance

$$\left(2\frac{\partial}{\partial r} - \frac{\partial^2}{\partial r^2}\right)(\eta_1\psi_1 - \eta_2\psi_2) + \sin\theta\,\frac{\partial\sigma}{\partial\theta} = 0 \qquad \text{at } r = 1. \tag{17}$$

When the Marangoni effect acts, the velocity field cannot be treated separately from the temperature and/or concentration fields. As mentioned earlier, here we consider the effect of radial temperature and/or concentration profiles. The results for this case may be represented in a quite general form, irrespective of what particular physico-chemical processes lead to these profiles.

For low Peclet numbers, in the leading approximation we have just unperturbed radial distributions of temperature and/or concentration, and hence homogeneity along the drop surface. A non-trivial effect on the tangential stresses (17) appears only in the next-order approximation, when a convective contribution to the temperature and/or concentration fields breaks the spherical symmetry. The temperature and/or concentration variation along the surface is a linear combination of the constants occurring in (15). The same is the case for the surface tension if we assume that it changes linearly with temperature and/or concentration.

In each particular problem, a (suitably modified) Marangoni number m is introduced, which is proportional to the surface tension variation with temperature or concentration and to a characteristic radial temperature or concentration gradient. Then condition (17) yields a set of equations corresponding to different modes which correspond to different orders of the Gegenbauer polynomials:

$$\left[\phi_1(\{P\})\,m + 1 + 3\beta/2\right] U_\infty + [\phi_2(\{P\})\,m + 1 + \beta]\,A_2 = 0 \tag{18}$$

$$(m - m_n)A_n = 0 \qquad (n = 3, 4, \ldots) \tag{19}$$

where $\beta = \eta_2/\eta_1$ is the ratio of dynamic viscosities; $\phi_1(\{P\})$ and $\phi_2(\{P\})$ are some functions of a set of material parameters $\{P\}$; m_n are also functions of the material parameters of the problem. Note that equations (19) give $A_n = 0$ for $m \neq m_n$ and leave A_n free for $m = m_n$. Note also that in the absence of the Marangoni effect ($m = 0$) equations (16), (18) provide exactly the Rybczynskii–Hadamard expression (4) for the drag force F_D.

In general, the expressions within square brackets in (18) can be either positive or negative, and hence A_2/U_∞ can be either negative or positive. Consequently, the force (16) can be either drag (U_∞ and F_{hd} have the same sign) or thrust (U_∞ and F_{hd} have opposite signs). The origin of this thrust has already been explained in section 2 using the example of a drop with uniform internal heat generation placed in a liquid with a uniform temperature distribution [33]. Here to complete our arguments let us consider another example, namely, a drop suspended in a homogeneous solution of a surfactant which is adsorbed at the drop surface and then disappears there in a first-order chemical reaction. In this case the Marangoni stresses are created by the variation in the surfactant concentration at the surface of the drop. To reduce the drag down to zero or even to create thrust we need a mechanism for the concentration at the leading pole of the drop to be higher than that at the trailing one. The surface chemical reaction does provide such a mechanism. As we consider the case of surfactant consumption, the concentration at the surface is lower than the homogeneous value at infinity. Thus the flow brings to the leading pole a higher concentration of surfactant. Generally, there is competition between this and other mechanisms (for example, convective transport of the surfactant along the surface film to the trailing pole), and eventually the possibility of reducing the drag and even of obtaining thrust [25].

There are two *critical* values m_1 and m_2 of the parameter m at which the coefficient A_2 vanishes and diverges to infinity, respectively (for $U_\infty \neq 0$). Equation (18) gives

$$m_1 = -(1 + 3\beta/2)/\phi_1$$
$$m_2 = -(1 + \beta)/\phi_2.$$

Thus we see that when $m = m_1$, according to linear theory the drop experiences no drag, and its velocity diverges to infinity irrespective of the value of the buoyancy. If, on the other hand, $m = m_2$, the drop experiences unbounded drag/thrust, whatever the value of its velocity may be. These two results show the limitation of the linear study in some asymptotically small vicinities of m_1 and m_2. Thus corrections with higher-order contributions must be taken into account (a weakly non-linear analysis for these cases is provided later on). Nevertheless, the conclusion that when m is close to m_1 (m_2) the drop motion induced by buoyancy is faster (slower) than the corresponding motion in the absence of the Marangoni effect can already be reached on the basis of the linear analysis.

In view of the similarity of the linear problem and the problem for the neutral steady perturbations, one can see that in the absence of buoyancy the critical values m_n ($n = 1, 2, \ldots$) of the Marangoni parameter m correspond to the neutral stability of the motionless state of the drop and of inner and outer fluids. Each m_n ($n = 1, 2, \ldots$) represents the steady instability threshold for the corresponding mode of (15). For the higher-order modes ($n = 3, 4, \ldots$), which describe the motion of the fluids inside and outside the drop without its translation, this conclusion follows directly from (19). The critical value m_1 is the instability threshold for drop translations. Indeed, in the absence of buoyancy the demand that the net force acting on the drop vanishes reduces to that of having a vanishing hydrodynamical force (16). As at $m = m_1$ we have $A_2 = 0$, this condition is satisfied for an arbitrary velocity U_∞.

The interpretation of the critical value m_2 is less obvious. It is the instability threshold for a *fixed* drop, i.e. a drop which is forced to remain at rest as a whole whatever flows develop around it, in contrast to a *free* drop, which is free to translate. Indeed (18) gives arbitrary A_2 for $m = m_2$, $U_\infty = 0$. Later on we consider the concepts of free and fixed drops with further generality. That is, we shall call a drop moving under a given buoyancy force *free*, while a drop driven at a given velocity will be called *fixed*.

As far as the stationary situation is concerned it makes no difference whether the drop is free or fixed. The difference appears only for the evolution problem—in particular, for linear stability. Note that our problem, when naturally extrapolated to the transient situation, corresponds in fact to a free drop. In order to consider the case of a fixed drop one has to incorporate some mechanism capable of keeping the drop at a fixed velocity whatever flows develop outside and inside it. Thus when we speak of a fixed drop, we tacitly assume the availability of such a mechanism.

While the instability thresholds for free and fixed drops are different—m_1 and m_2 respectively—those for the higher-order modes m_n ($n = 3, 4, \ldots$) do not depend on whether the drop is free or fixed. From now on here we deal only with the case of a free drop. Note that the m_n ($n = 1, 2, \ldots$) are not necessarily associated with the axisymmetrical modes. In view of the spherical symmetry of the motionless base state this result is enough to ensure that a three-dimensional analysis will not add new thresholds.

In the simplest cases all m_n ($n = 1, 2, \ldots$) have the same sign and the subset $|m_n|$ gets higher values as n becomes higher [25, 28, 33]. Nevertheless in the general case one may expect that the thresholds m_n can be ordered in any sequence. The number n of the lowest positive threshold and that of the highest negative one, if any, determine the mode(s) for which the Marangoni instability occurs first as $|m|$ is increased. For the simplest cases the translational instability appears first.

6. Instability and non-linear drop motions

6.1. The case where $m \to m_1$; autonomous motion

In this section we consider m close to the translational instability threshold m_1. Thus we assume that $|m - m_1| \ll 1$, $|m - m_n| \simeq 1$ ($n = 2, 3, \ldots$). In the linear approximation, higher-order terms in low Reynolds/Peclet numbers have been neglected in equation (18). However, if the coefficient $m - m_1$ is small enough such a limitation may not be adequate. Thus to discuss this case some higher-order contributions need to be taken into account in equation (18). It is also clear that the higher-order terms to be incorporated must depend on U_∞ but not on A_2 since in the latter case they would be only a small correction to the second term in the left-hand side of equation (18). Besides, symmetry demands that the additional contribution is odd in U_∞. Finally, instead of equation (18) we have

$$\phi_1(m - m_1)\, U_\infty + K_1 \frac{a}{d}\, |U_\infty|\, U_\infty + \phi_2\, (m - m_2)\, A_2 = 0 \tag{20}$$

where d is a mass/thermal diffusivity coefficient specific to each particular problem.

When $\phi_1(m - m_1)K_1 < 0$, equation (20) has three solutions for U_∞ if the force (or A_2 which is related to the force by (16)) is weak enough. This corresponds to three different regimes of drop motion. The dependence of F_{hd} on U_∞ is defined by (20), (16). In the absence of buoyancy ($A_2 = 0$), besides the motionless state we may have autonomous or self-sustained drop motion at a velocity whose absolute value is

$$|U_{aut}| = -\frac{\phi_1\, (m - m_1)\, d}{K_1\, a}.$$

Depending on the signs of ϕ_1 and K_1, this motion is either supercritical (i.e. for $|m| > |m_1|$) or subcritical. As was already said, in the absence of buoyancy all directions in space are allowed for the autonomous motion and the breakdown of this symmetry can only be induced by the initial conditions.

A stability analysis restricted to steady disturbances [25] shows that the states corresponding to the extrema of the force versus velocity curve are neutrally stable to velocity perturbations aligned with the base velocity, while the autonomous motion regime is neutrally stable to orthogonal perturbations. There are no other neutrally stable states among those described by equation (20). Then, in principle, if we knew the stability status of the motionless regime ($U_\infty = 0$), and if we were sure that oscillatory instability does not occur in our system, we could extrapolate by continuity the stability status of the state of the remaining motion regimes. For example, if the motionless state of the drop (in the absence of buoyancy) is unstable, we could conclude that the motion regimes with velocities in the range $0 < |U_\infty| < |U_{aut}|/2$, where $|U_{aut}|/2$ corresponds to the extrema of the curve of F_{hd} versus U_∞, are unstable to both parallel and orthogonal perturbations. The motions with $|U_{aut}|/2 < |U_\infty| < |U_{aut}|$ become stable to parallel perturbations, albeit still being unstable to orthogonal ones. For $|U_\infty| > |U_{aut}|$ the motion is stable. Alternatively, if the motionless regime is stable, in the first of the above velocity intervals we have stability, in the second one, instability to parallel perturbations and stability to orthogonal ones, while in the third interval, we have instability to both types of perturbation.

If an oscillatory instability is possible, the above argument does not hold. A sufficient condition for overstability could be e.g. that the rest state is unstable to steady disturbances *in the subcritical region*, i.e. for $|m| < |m_1|$. This follows from two facts: (1) at $m = 0$ there is stability, (2) m_1 is the only possible threshold for the steady instability. Then, by continuity, the oscillatory instability should inevitably exist with a threshold below m_1, the threshold for the steady instability. A detailed study of this possibility is under way and will be published elsewhere.

Note that to respect symmetry in the Landau theory, a weakly non-linear analysis usually involves a cubic non-linear term. However, equation (20) contains a quadratic term with the symmetry taken into account by writing it as a pseudoquadratic term. This remarkable difference originates in the specific properties of the matching problem at low Peclet number for the temperature/concentration field when the form of the non-linear term of equation (20) is determined by the region far away from the drop [33]. Then one may expect that for relatively high diffusivity coefficients of the outer fluid relative to that of the inner fluid the usual cubic term would dominate.

6.2. *The case where $m \to m_2$; non-single-valued hydrodynamical force*

Let us consider now the case where $m \to m_2$, $|m - m_1| \simeq 1$, $|m - m_n| \simeq 1$ ($n = 3, 4, \ldots$). Equation (18) must be modified with a non-linear term depending on A_2 which has to be odd in A_2 by symmetry. We have

$$\phi_1\,(m - m_1)\,U_\infty + \phi_2\,(m - m_2)\,A_2 + K_2\,\frac{a^2}{d^2}\,A_2^3 = 0 \qquad (21)$$

where K_2 is a coefficient depending on the material parameters of the problem (see [25] for the particular expression for K_2 corresponding to the case of a surfactant reacting at the drop surface). The hydrodynamical force versus the velocity U_∞ can be obtained using (16), (21). For a given low enough drop velocity, three possible values of the force are available—hence a non-single-valued hydrodynamical force. Thus three different levels of buoyancy can lead to the same speed of the drop. A remarkable particular case corresponds to a *levitating* drop even in the presence of buoyancy. A stability analysis [25] predicts stability for all of the regimes found above if m_2 is minimal in absolute value for all m_n ($n = 1, 2, \ldots$) having the same sign as m_2.

6.3. Higher-order modes

Consider the higher-order modes ($n = 3, 4, \ldots$). Symmetry demands that the amplitude equations for the odd-number modes and for the even ones be different. Actually, the even modes are symmetric: changing the sign of the amplitudes $A_4, A_6, \ldots$ in (15) leads to the same motion, but differently oriented in space. By rotating the coordinate system by π we come back to the initial form. As for the odd modes, changing the sign of the amplitudes $A_3, A_5, \ldots$ corresponds to a different flow pattern which cannot be obtained from the initial form by rotation. Symmetry determines the form of the amplitude equations, which are

$$(m - m_n)A_n + K_n \frac{a}{d} A_n^2 = 0 \tag{22}$$

for the odd-number modes ($n = 3, 5, \ldots$), and

$$(m - m_n)A_n + K_n \frac{a^2}{d^2} A_n^3 = 0 \tag{23}$$

for the even-number modes ($n = 4, 6, \ldots$). Here K_n are again constants that depend on the material properties of the system. Note that a non-zero solution of equation (22) exists for both $m - m_n < 0$ and $m - m_n > 0$ (both subcritically and supercritically), while for the symmetric mode (equation (23)) it exists only under one of these conditions, depending on the sign of K_n.

7. Concluding remarks

Drops and bubbles are ubiquitous. Stokes, Rybczynskii, Hadamard, Boussinesq and others provided the drag law experienced by a drop. Young, Goldstein and Block and others studied the motion due to interfacial stresses induced on a drop immersed in a medium where an imposed thermal gradient exists. They showed the possibility of its levitation in the presence of gravity. Levich, Sanfeld and others pointed out the role played by surfactants in affecting the drag law and the possible fluid motion inside the drop. After providing a succinct account of the results of the earlier theories and some relevant experiments, we have shown that when there is heat and/or mass transfer between a drop and its environment, a chemical reaction at the surface of a drop or internal heat generation, coupled to the Marangoni effect, i.e. the surface stresses and subsequent flow induced by the surface tension gradients, such *activity* and coupling lead to remarkable consequences. For instance, we have that (i) the drag on the drop may actually become thrust, and hence self-propulsion due to instability of the motionless state, (ii) autonomous motion may offer a multiplicity of steady values for a given external (weak) force like buoyancy, and (iii) levitation is possible for multiple buoyancy levels.

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