ARTICLES

A swarm of Stokeslets with interfacial tension

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A formal analogy between sedimenting drops in Stokes flow and a swarm of Stokeslets [Machu *et al.*, J. Fluid Mech. (in press)] is extended to include interfacial tension. Using a cohesive potential, mean curvature is extended as a meaningful quantity off the interface, allowing the boundary-integral formulation to be rewritten in volumetric form. A prescription for assigning forces to the Stokeslets comprising the swarm incorporates the action of interfacial tension without having to identify a boundary surface. Numerical simulations agree with linear small-deformation theory, and reproduce the spontaneous coalescense of two touching drops. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1369124]

I. INTRODUCTION

For one or more sedimenting drops with the same viscosity as the surrounding liquid and vanishing interfacial tension, a previous paper[1] has used a fundamental analogy with a swarm of Stokeslets to calculate coalescense and break-up phenomena involving extreme distortion and intertwining of the interfaces. The purpose of this paper is to show how a representation of interfacial tension can be built into the swarm analogy, thereby bringing a wide range of fully three-dimensional free-surface problems within reach of a simple, conceptually appealing method of computer simulation.

As is described in textbooks and monographs on interfacial phenomena and surface chemistry,[2–4] the idea that molecules tend to lower their collective potential-energy by agglomerating so as to be surrounded as completely as possible by their nearest-neighbors leads naturally to the standard concept of surface energy: work required to increase the number of molecules residing in an incompletely surrounded state at the surface. What seems to be missing is a concise line of argument leading directly from this basic idea to the mean curvature term (which can be regarded as a dynamic version of the Young–Laplace equation) in the boundary-integral equation of creeping flow.[5–7] The key to making this link is to abandon the conceptual detour implicit in the continuum representation of interfacial tension as a property of the *interface* and return to the molecular origins—albeit in a stylized form. Previous volumetric formulations of interfacial tension[8–13] have led to fairly complicated formulas be-

cause they essentially only *convert* what is defined artificially as a surface property into volumetric form. These models will be discussed in detail at the end of Sec. II. Our greatly streamlined procedure assigns to each Stokeslet in the swarm an appropriate force vector that—in collective terms—mimics the action of interfacial tension, without ever having to identify an interfacial boundary.

II. CONTINUUM FORMULATION

We begin with the well-known boundary-integral formulation for gravitationally driven Stokes flow of one or more drops[6,7,14–16] and argue backwards toward a stylized molecular view. For the case of equal viscosities of the drop(s) and surrounding liquid, the velocity field throughout both phases and the interface is given (in dimensionless form) by

$$u(r,t) = \int_{\mathcal{S}(t)} \{ \mathcal{B}e_g \cdot q - 2H(q,t) \} n[q] \cdot G(q-r) \, d\mathcal{S}[q], \tag{1}$$

with G the point-force solution (Stokeslet)

$$G(r) = \frac{1}{8\pi r}(I + \hat{r}\hat{r}), \quad \hat{r} = r^{-1}r, \quad r = \|r\|, \tag{2}$$

H the local mean curvature of the interface, and e_g a unit vector pointing in the direction of gravity. In Eq. (1) the position r is fixed, while the dummy argument of integration q runs over the surface $\mathcal{S}(t)$. Dimensionless velocities are referred to the interfacial-viscous scale σ/μ, with σ the interfacial tension and μ the viscosity of both phases. The strength of gravitational forces (which tend to deform the drop) compared with interfacial tension (which tends to restore a spherical shape) is indicated by the Bond number

$$\mathcal{B} = \frac{(\Delta\rho)g a^2}{\sigma}, \tag{3}$$

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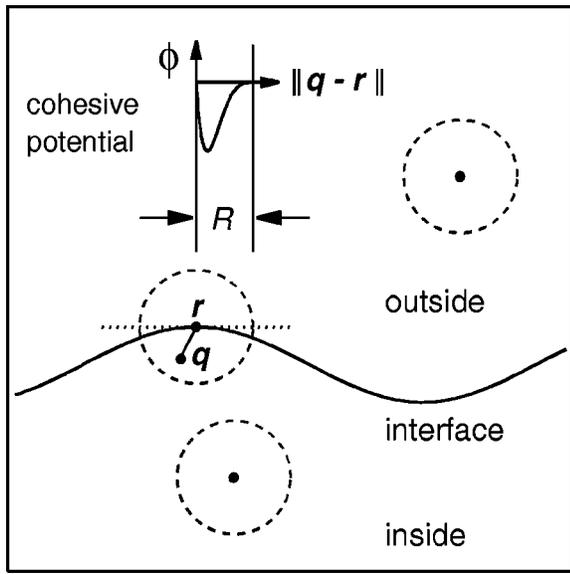[b)]Institut für Strömungslehre und Wärmeübertragung, Technische Universität Graz, Graz, Austria.

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FIG. 1. Approximation of the mean curvature of an interface using a short-range cohesive energy that is defined at *all* points r, not just those lying on the surface.

with a the radius of the (undeformed) drop, $\Delta\rho$ the density difference between the drop and surrounding liquid, and g the acceleration of gravity. The gravitational term can be converted into a volume integral using the divergence theorem, because the scalar quantity being integrated over the surface ($e_g \cdot r$) is obviously defined at *all* points.[1] But there seems at first to be no simple, general way to extend the mean curvature as a meaningful quantity off the interface. If, however, we are willing to deal only with an approximation H^* instead of H itself, then there is hope for a volumetric transformation of the interfacial tension term as well.

To approximate the mean curvature we shall devise the concept of an energy ''probe'' that responds to the degree to which it is surrounded by (material) points associated with the drop phase (Fig. 1). We define a radially symmetric short-range potential $\Phi(r)$ using a scalar function $\phi(x)$ of dimensionless distance $x \geq 0$ that is continuous and piecewise smooth, and vanishes for all $x \geq 1$. From this follows the total energy of interaction $\Psi(r,t)$ of the probe point r with all those points q of the drop phase $\mathcal{V}(t)$ that also lie within a spherical test volume $\mathcal{T}(r;R)$ of radius R centered at r

$$\Psi(r,t) = \int_{\mathcal{V}(t) \cap \mathcal{T}(r;R)} \Phi(q-r) \, d\mathcal{V}[q]$$

$$= \int_{\mathcal{V}(t) \cap \mathcal{T}(r;R)} \phi\left(\frac{\|q-r\|}{R}\right) d\mathcal{V}[q]. \quad (4)$$

This energy is defined for *all* points r, and has the useful property that, when r happens to lie on the surface of the drop, it yields an approximation of the local mean curvature

$$H^*(r,t) \stackrel{\text{def}}{=} \frac{2I_2}{RI_3} - \frac{\Psi(r,t)}{\pi R^4 I_3} = \begin{cases} H(r,t) + O(R^2), & r \in \mathcal{S}(t), \\ \text{well defined}, & \text{otherwise}, \end{cases} \quad (5)$$

with

$$I_n = \int_0^\infty \phi(x) x^n \, dx = \int_0^1 \phi(x) x^n \, dx. \quad (6)$$

With reference to a local Cartesian coordinate system centered at the surface point r, Eq. (5) is obtained by integrating ϕ over that portion of the spherical sampling volume $x^2 + y^2 + z^2 < R^2$ that lies between the interface $z = f(x,y)$ and the tangent plane $z = 0$. The mean curvature comes from the quadratic terms in a Taylor expansion of $f(x,y)$.

The potential probe acts like a ''filter'' that discards surface details below the lengthscale R. In addition to the requirements mentioned before Eq. (4), we should choose $\phi(x)$ so that neither I_2 nor I_3 vanish—the latter to avoid zero in the denominator of Eq. (5); the former so that agglomeration of particles has a net energetic effect. Otherwise $\phi(x)$ is arbitrary; it could (but need not) mimic an actual intermolecular potential.

Now we can apply the divergence theorem also to the interfacial tension term in Eq. (1) to obtain the volumetric form

$$u(r,t) = \int_{\mathcal{V}(t)} \{\mathcal{B}e_g - 2\nabla H^*(q,t)\} \cdot G(q-r) \, d\mathcal{V}[q]. \quad (7)$$

The quantity ∇H^* has an appealing physical interpretation as the cohesive force exerted upon the material point r by nearby material points q in the drop

$$\nabla H^*(r,t) = \frac{-1}{\pi R^5 I_3} \int_{\mathcal{V}(t) \cap \mathcal{T}(r;R)} \phi'\left(\frac{\|q-r\|}{R}\right) \left\{\frac{q-r}{\|q-r\|}\right\} d\mathcal{V}[q]. \quad (8)$$

The imbalance of this force for molecules lying close to the surface represents interfacial tension without having to deal with differential geometry of the surface.

Previous volumetric formulations of interfacial tension—used in volume-of-fluid (VOF) methods[13] and smoothed particle hydrodynamics (SPH)[12]—are more complicated than Eqs. (5) and (8). These formulations can be divided into two main categories. The first approach[8,10] uses a standard expression for mean curvature in terms of the unit normal vector n on the surface

$$H = -\nabla_s \cdot n \quad \text{or} \quad -\nabla \cdot n, \quad (9)$$

and extracts n from a piecewise-constant phase function F (also referred to as a ''color function'') that indicates the shape of the drop

$$n = \frac{\nabla F}{\|\nabla F\|}, \quad F(r) = \begin{cases} 1, & r \in \mathcal{V} \\ 0, & \text{otherwise}. \end{cases} \quad (10)$$

For numerical purposes the infinitesimally thin interface can be smeared out. Typically this is done with a weight function or mollifier, which is essentially a suitably normalized version of the potential function Φ from Eq. (4). The smoothed phase function is equivalent to our potential function Ψ. Thus the mean curvature is estimated as follows:

$$H \approx -\nabla \cdot \left(\frac{\nabla \Psi}{\|\nabla \Psi\|}\right). \quad (11)$$

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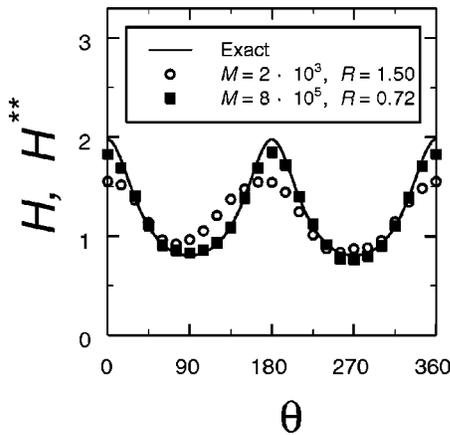Stokeslets with interfacial tension   1551

FIG. 2. Approximate vs exact mean curvature of a prolate spheroid. The major (x) axis is $a=(5/3)^{2/3}\approx1.406$, and the minor ($y,z$) axis is $b=(3/5)^{1/3}\approx0.843$. This gives the same volume as the unit sphere, with axis ratio 0.6. We plot the exact curvature H and its numerical approximation H^{**} from Eq. (15) against polar angle θ (measured counterclockwise from the x axis) along the elliptical trace of the surface in the (x,y) plane. Two random discretizations are used to compute H^{**}: $M=2000$ with $R=1.5$ vs $M=800\,000$ with $R=0.72$.

For comparison, Eq. (5) can be cast in the form $H\approx\mathcal{C}_1-\mathcal{C}_2\Psi$, which saves two orders of differentiation and the attendant numerical inaccuracies.

The second kind of volumetric formulation[9,11] starts from a capillary pressure tensor

$$\mathbf{T}=-\sigma(\mathbf{I}-\mathbf{nn})\|\nabla F\|, \tag{12}$$

where $\|\nabla F\|$ operates like a surface delta function to pick out the interface. The volumetric force attributed to capillarity is then

$$\mathbf{f}_s=-\nabla\cdot\mathbf{T}=\nabla\cdot\left[\sigma\left(\|\nabla F\|\mathbf{I}-\frac{(\nabla F)(\nabla F)}{\|\nabla F\|}\right)\right]. \tag{13}$$

Again, the smoothed phase function Ψ may replace F in numerics. This formulation has been interpreted physcally in terms of a surface excess pressure.[9] Our much simpler Eq. (8) saves one order of differention ($\mathbf{f}_s\approx\mathcal{C}\nabla\Psi$), and expresses intermolecular cohesion directly.

III. A SWARM OF STOKESLETS

For numerical purposes the volume integral (7) is discretized with a Monte Carlo procedure, whereby M Stokeslets are placed in a statistically uniform, random arrangement $\mathbf{r}_1,\ldots,\mathbf{r}_M$ throughout the drop phase. To each point we attribute the volume element $\delta V=V/M$, and the integral is replaced by a summation

$$\mathbf{u}_m^{**}(\mathbf{r}_1,\ldots,\mathbf{r}_M)=\sum_{n\neq m}(\delta V)\mathbf{f}_n\cdot\mathbf{G}(\mathbf{r}_n-\mathbf{r}_m). \tag{14}$$

The resulting statistical error is of order $(\delta V)^{1/2}\ln(\delta V)$.[17] Based upon the same statistical quadrature, Fig. 2 shows numerical results for the approximate mean curvature

$$H^{**}(\mathbf{r})\overset{\text{def}}{=}\frac{2I_2}{RI_3}-\frac{\delta V}{\pi R^4I_3}\sum_{m=1}^{M}\phi\left(\frac{\|\mathbf{r}_m-\mathbf{r}\|}{R}\right), \tag{15}$$

obtained using a dimensionless potential

$$\phi(x)=\begin{cases}(1-x)^{3p}-(1-x)^3, & 0\leqslant x\leqslant1\\0, & x>1\end{cases}, \tag{16}$$

where the exponent p is chosen (by iterative solution) to match the minumum of the potential well (see Fig. 1) with the mean interparticle spacing $(\delta V)^{1/3}$. The forces in Eq. (14) are given by the discrete version of Eq. (8)

$$\mathbf{f}_m(\mathbf{r}_1,\ldots,\mathbf{r}_M)=\mathcal{B}\mathbf{e}_g-\frac{2(\delta V)}{\pi R^5I_3}\sum_{n\neq m}\phi'\left(\frac{\|\mathbf{r}_n-\mathbf{r}_m\|}{R}\right)\left\{\frac{\mathbf{r}_n-\mathbf{r}_m}{\|\mathbf{r}_n-\mathbf{r}_m\|}\right\}. \tag{17}$$

In principle, some provision may be required to ameliorate the singularity of the Stokeslet in Eq. (14): otherwise an unphysically large velocity can result if two Stokeslets happen to lie too close together[1,18]—which is not prevented by the dynamic equations and could enter simply through the initial configuration of the swarm.

The above scheme requires two refinements. First, local statistical fluctuations in the forces from Eq. (17) should be smoothed out before using them in Eq. (14), in order to quell a kind of spurious numerical dispersion. We found five successive weight-function averages, with an active radius of $5(\delta V)^{1/3}$, to be sufficient for this purpose. The analogous numerical artifact of ''parasitic currents'' arising in VOF methods has been dealt with in a related way.[9,11]

The second refinement concerns a numerical drift effect that causes the Stokeslet clouds to expand unphysically. It seems that neglected terms hidden in the instantaneous discretization error need not cancel out when summed all together, and can accumulate to a significant volume over a sufficiently long time (as with a nonconservative finite-difference formulation for a parabolic PDE). This numerical artifact can be counteracted by retaining the $O[(\delta V)^{2/3}]$ slip velocity between each Stokeslet and the local fluid velocity vector

$$\frac{d\mathbf{r}_m}{dt}=\mathbf{u}_m^{**}(\mathbf{r}_1,\ldots,\mathbf{r}_M)+\alpha(\delta V)^{2/3}\mathbf{f}_m, \tag{18}$$

which is formally negligible at each instant but numerically important on a cumulative basis. Here the particulate character of the method asserts itself. The correction term is also equivalent to an analytical evaluation of the weakly singular kernel over a particular choice for the volume element surrounding each point force. We regard α as an adjustable renormalization parameter, and find that setting $\alpha=(6\pi)^{-1}$ yields good results for conservation of volume—over durations long enough to track relaxation and coalescense of drops from beginning to end. (We have also tried an adaptive scheme for adjusting α at each time step based upon an estimated rate of expansion; this procedure will be described in a separate paper.)

IV. NUMERICAL RESULTS FOR THREE TEST PROBLEMS

A useful conclusion in implementing Eqs. (14), (17), and (18) is that even a coarse discretization (in terms of R and M) gives good results, provided that we smooth the cohesive
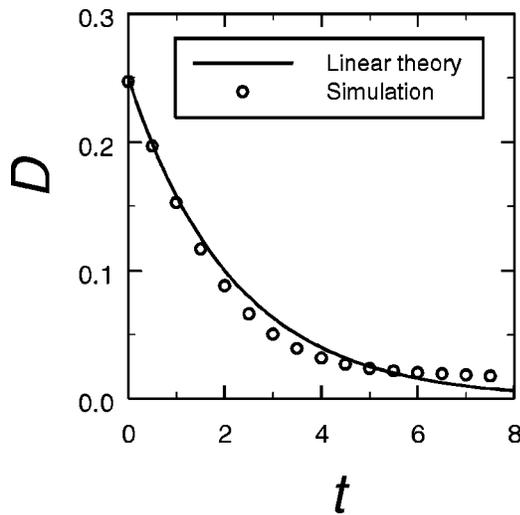
FIG. 3. Deformation D as a function of time t for relaxation of the spheroidal drop described in Fig. 2 to a spherical shape in otherwise quiescent liquid. Note good agreement with the exponential decay predicted by small-deformation theory.

FIG. 5. Shape evolution of sedimenting drops, with gravity pulling toward the right. (a) Initial stages in the formation of a torus (and subsequent break-up) of an initially hemispherical drop in the absence of interfacial tension. (b) At unit Bond number the shape instability is counteracted by interfacial tension, which restores a spherical shape.

forces. All simulations were carried out with 2000 Stokeslets per drop (in the initial configuration), $R=1.5$ for the radius of cohesion, and a fourth-order Runge–Kutta scheme for integrating the coupled, nonlinear ODE's using the time step $\delta t=0.1$. In practice we did not encounter problems with the singularity of the Stokeslet, and saw no need to modify it.[1]

Applied to the relaxation of a spheroidal drop in quiescent liquid, the classical, linear small-deformation theory[6,19] gives an exponential decay with time of the deformation pa-
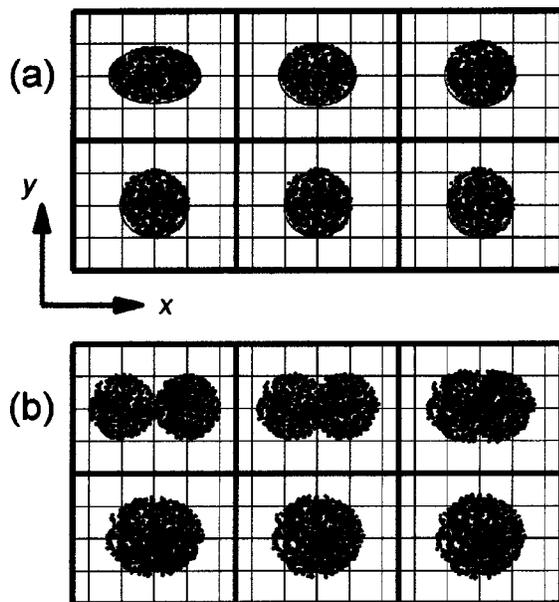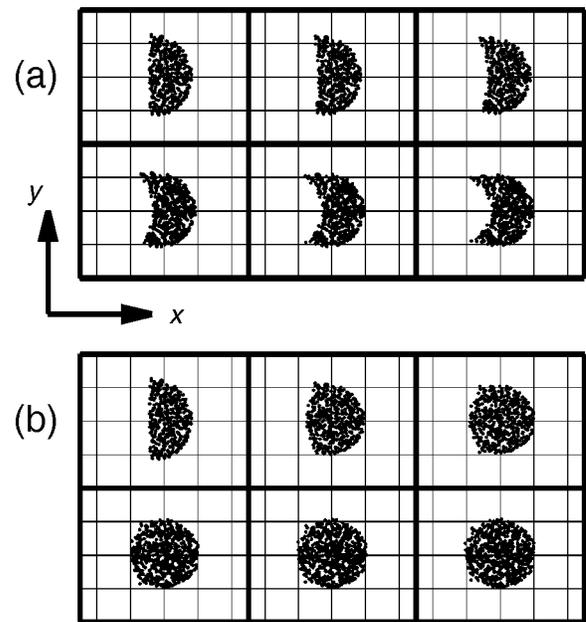

FIG. 4. Simulated shape evolution of drops driven by surface tension in otherwise quiescent liquid: (a) relaxation of the initially spheroidal drop from Fig. 3; (b) spontaneous coalescense of two initially touching drops. The dimensionless lengthscale is set visually by a square grid of unit spacing. A meridional view in the (x,y) plane is approximated by showing only those Stokeslet positions lying between $z=\pm0.2$. The time interval between successive snapshots is $\Delta t=1.5$. For part (a) the elliptical outlines from small-deformation theory are also shown.

rameter: $D(t)=(a-b)/(a+b)=\exp(-16t/35)$, where a and b are the longitudinal and transverse axes, respectively. Here we estimate a and b for the swarm by matching a spheroid to the second-moment tensor of the distribution of points. For an initially prolate drop, $D(0)=0.25$, of the same volume as the unit sphere, Fig. 3 shows good agreement between this asymptotic formula and our simulation. Six snapshots from the shape evolution appear in Fig. 4(a), at equal intervals for $0\leqslant t\leqslant7.5$. For comparison, the elliptical outline from small-deformation theory is also plotted in each case. The coalescense of two initially touching drops is simulated in Fig. 4(b). Because of the large radius of cohesion, two drops that start off with an appreciable gap will also be drawn together and coalesce. We are currently studying how to model the retarding effect of film drainage.[15,20–22] Finally, Fig. 5 shows how interfacial tension moderates the shape instability of a sedimenting drop. Our previous paper[1] identified the hemisphere as one among several shapes that, at infinite Bond number, undergo a process of torus formation and subsequent breakup. The initial stages of this shape evolution are depicted in Fig. 5(a). At unit Bond number this process is inhibited, and the drop "snaps back" to a spherical shape. For the relaxing spheroid [Fig. 4(a)] the second-moment scheme yielded the approximate volume of the swarm throughout the simulation. For Figs. 4(b) and 5(b) there was no analytical shape formula to fit to the cloud at intermediate times. But we could at least check the final equilibrium, which had to be the sphere. By these estimates, volume was conserved well within 2%.

V. CONCLUDING REMARKS

The essential advance reported in this paper has been the extension of our particulate method for gravity-driven mo-

tion and deformation of drops with the same viscosity as the surrounding liquid[1] to include cohesion within the drop phase. Thereby we model the *action* of interfacial tension, without any explicit treatment of the (fuzzy) boundary of the swarm of Stokeslets. Other volumetric formulations of the capillary forces[8–13] are physically less direct and mathematically more complicated. Surprisingly coarse clouds of Stokeslets give accurate results for a classical test problem (relaxation of a prolate spheroid) and even capture the spontaneous coalescence of two initially touching drops. We attribute this robustness of our numerical scheme to its basis in the underlying substructural physics. That is the crucial ingredient and strength of the approach; there seems to be much latitude in the details of implementation.

Perhaps the greatest potential of the technique advanced in this paper lies in local augmentation of boundary-element methods (BEM) or other established numerical techniques, in (often temporary) regions of problematic fine structure, such as interfacial coalescence and rupture. Linearity of the Stokes equations would make it straightforward, by simple superposition, to merge the robustness of Stokeslet clouds with the elegant efficiency of BEM.

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