Effective surface shear viscosity of an incompressible particle-laden fluid interface

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Effective surface-shear viscosity of an incompressible particle-laden fluid interface

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The presence of even a small amount of surfactant at the particle-laden fluid interface subjected to shear makes surface flow incompressible if the shear rate is small enough [T. M. Fischer *et al*., J. Fluid Mech. **558**, 451 (2006)]. In the present paper the effective surface shear viscosity of a flat, low-concentration, particle-laden incompressible interface separating two immiscible fluids is calculated. The resulting value is found to be 7.6% larger than the value obtained without account for surface incompressibility.

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I. INTRODUCTION

To minimize total interfacial energy, suspended particles can self-assemble on the interface between two fluids [1]. The structure and the dynamics of the particles at the interface are controlled by their interaction, both direct (e.g., capillary, steric, electrostatic, magnetic, van der Waals) and mediated by the surrounding fluids (hydrodynamic), as well as by size and chemical composition of the adsorbed particles. Both the structure and the dynamics of the particles contribute to their ability to stabilize particle-laden films, foams, and emulsions, and to control the transfer of matter through the interface [2–4].

At macroscopic scales, the particle-laden fluid interface can be viewed as a continuous infinitely thin fluid interface with some *effective* static and dynamic properties, which can be measured experimentally. Examples of static effective properties of particle-laden fluid interfaces are surface tension [5], bending [6], and saddle-splay [7] elastic moduli. Dynamic properties include, in particular, surface rheological parameters [4].

The study of the surface rheology of particle-laden fluid interfaces can provide new insight into their structure and properties. At the same time, rheology of these systems poses many experimental and theoretical challenges. This makes it desirable constructing and investigating "model interfaces," which could be utilized as reference systems in well-defined limits, such as the extremes of a purely Newtonian and purely elastic interface.

The rheology of a particle-laden interface becomes purely Newtonian when direct interparticle interactions are small, which is the case when adsorbed particles are separated widely enough. In this case effective surface shear and dilatational viscosities arise due to viscous energy dissipation in the surrounding bulk fluids. They can be calculated analytically for the special case of the spherical particles having a solid-fluid contact angle (between particles' surface and fluid-fluid interface) $\pi/2$, neglecting surface viscosity [8,9]. The results can be used as a starting approximation for more complicated systems in which the interparticle interactions of a different nature cannot be neglected.

The presence of adsorbed surfactant film on an interface introduces additional viscoelastic stresses. If the concentration of surfactant is small, additional surface viscosity and elasticity can be neglected. However, at small concentrations this case does not reduce to the case of surfactant-free interface. The presence of even small amount of surfactant results in effective surface incompressibility if the shear rate is small enough [10,11].

The effect of the incompressibility of the surface flow upon the effective surface shear viscosity of a low-concentration particle-laden interface is calculated in this paper.

II. MODEL

We consider the steady flow of a system of identical rigid spherical particles of radius R adsorbed at the flat interface between two incompressible fluids. We neglect gravity and assume the interfacial tensions favor a contact angle $\pi/2$, so that the particles are located with the equator coinciding with the interfacial plane.

We suppose the interface to be macroscopically thin, having surface tension high enough to keep interface flat in the flow, and incompressible. These conditions correspond to small shear rates which satisfy inequality Eqs. (40) and (41), respectively.

We assume surface concentration of the adsorbed particles,

$$\phi = \frac{\pi R^2 N}{A}, \tag{1}$$

with N being the number of particle in area A, to be small, so that the motion of any particle is not affected by other particles.

The energy dissipation rate in this system has two contributions, dissipation in the bulk fluids, and the dissipation at the interface between two fluids. We shall neglect surface dissipation. This assumption can be expressed as Boussinesq number, defined as the ratio of surface viscosity of the interface to the viscosity of bulk fluids,

$$\mathrm{Bo} = \frac{\eta_s}{(\eta_1 + \eta_2)R}, \tag{2}$$

being small, $\mathrm{Bo} \ll 1$.

We assume both fluids 1 and 2 have the same densities, $\rho_1 = \rho_2 \equiv \rho$, and shear viscosities, $\eta_1 = \eta_2 \equiv \eta$. We write the equations of motion in a form valid for the entire fluid domain as

$$\nabla \cdot \boldsymbol{\sigma} = 0. \tag{3}$$

The stress tensor $\boldsymbol{\sigma}$ has bulk and interfacial contributions,

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}_b + \delta(z)\boldsymbol{\sigma}_s. \tag{4}$$

The bulk stress tensor,

$$\boldsymbol{\sigma}_b = -p\mathbf{I} + \eta[\nabla\mathbf{v} + (\nabla\mathbf{v})^{\mathrm{T}}], \tag{5}$$

where $\mathbf{v}$ is velocity, p is pressure, $\mathbf{I}$ is unit tensor, yields Stokes equations [12],

$$\eta\nabla^2\mathbf{v} = \nabla p, \tag{6}$$

$$\nabla\cdot\mathbf{v} = 0. \tag{7}$$

In Eq. (4), Dirac's delta function $\delta(z)$ is used to localize at the plane $z = 0$ the body force that arises from the jump conditions at the fluid interface (see Ref. [13] and references cited therein). We shall use the surface stress tensor in form

$$\boldsymbol{\sigma}_s = -\Pi\mathbf{I}_s, \tag{8}$$

Π being surface pressure, $\mathbf{I}_s = \mathbf{I} - \mathbf{nn}$ being surface projector, where we have neglected viscous contribution and dropped constant surface tension contribution since it does not produce any body forces for the flat interface. The tangential stress balance at the interface is

$$\mathbf{I}_s\cdot(\boldsymbol{\sigma}_2 - \boldsymbol{\sigma}_1)\cdot\mathbf{n} - \nabla_s\Pi = 0, \tag{9}$$

where $\nabla_s = \mathbf{I}_s\cdot\nabla$ is surface gradient operator.

The above formulation corresponds to no-slip boundary condition at the fluid interface. We further assume no-slip boundary condition at the surface of the particles. Surface continuity equation for incompressible interfacial flow reads

$$\nabla_s\cdot\mathbf{v}_s = 0. \tag{10}$$

Note that the component of the fluid velocity normal to the fixed interface should vanish,

$$\mathbf{n}\cdot\mathbf{v}_i = 0. \tag{11}$$

We shall consider the shear flow in the system with the motion of the fluid when unperturbed by both particles and a fluid interface being described by the velocity field

$$\mathbf{v}^{(0)} = \boldsymbol{\alpha}\cdot\mathbf{r}, \tag{12}$$

where the rate-of-strain tensor $\boldsymbol{\alpha}$ is symmetric, traceless, and constant. The applied rate-of-strain is supposed to be small and the unperturbed pressure is taken to be zero.

It is well known that a shear flow distribution may be decomposed into a symmetric shear and a rotation. By considering the motion in an appropriately chosen uniformly rotating rest frame, we need concern ourselves solely with the dissipation associated with a symmetric shear. Taking into account that the velocity at $z = 0$ is parallel to the interfacial plane XY, and both bulk and interfacial flow is incompressible, we can choose the symmetric rate-of-strain tensor to have the form

$$\boldsymbol{\alpha} = \begin{pmatrix} \alpha & 0 & 0 \\ 0 & -\alpha & 0 \\ 0 & 0 & 0 \end{pmatrix}. \tag{13}$$

The corresponding unperturbed velocity field is

$$v_x^{(0)} = \alpha x, \tag{14a}$$
$$v_y^{(0)} = -\alpha y, \tag{14b}$$
$$v_z^{(0)} = 0. \tag{14c}$$

The velocity, pressure, and surface pressure fields in presence of the particles and the fluid interface will be, respectively, written as

$$\mathbf{v} = \mathbf{v}^{(0)} + \mathbf{v}^{(1)}, \tag{15}$$

$$p = p^{(0)} + p^{(1)}, \tag{16}$$

$$\Pi = \Pi^{(0)} + \Pi^{(1)}. \tag{17}$$

The stress tensor can be decomposed as

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}^{(0)} + \boldsymbol{\sigma}^{(1)}, \tag{18}$$

where

$$\boldsymbol{\sigma}^{(0)} = -p^{(0)}\mathbf{I} + \eta[\nabla\mathbf{v}^{(0)} + (\nabla\mathbf{v}^{(0)})^{\mathrm{T}}] \tag{19}$$

is the stress tensor of unperturbed flow, and

$$\boldsymbol{\sigma}^{(1)} = \boldsymbol{\sigma}_b^{(1)} + \delta(z)\boldsymbol{\sigma}_s, \tag{20}$$

with

$$\boldsymbol{\sigma}_b^{(1)} = -p^{(1)}\mathbf{I} + \eta[\nabla\mathbf{v}^{(1)} + (\nabla\mathbf{v}^{(1)})^{\mathrm{T}}], \tag{21}$$

is the perturbation due to presence of particles and incompressible fluid interface.

III. EFFECTIVE SURFACE SHEAR VISCOSITY

In order to find an expression for the effective surface shear viscosity of the particle-laden interface, we shall compare expressions for the rate of viscous dissipation calculated in two ways. First, we consider the system as homogeneous, having an effective continuum interface with effective surface shear viscosity η_s^{eff}. Second, we consider the flow in presence of particles explicitly. Equating the energy dissipation rate in both cases will provide the expression for η_s^{eff}. In our derivation we shall closely follow the idea of Einstein for calculating the effective shear viscosity of a three-dimensional dilute suspension of solid spherical particles [14,15] in a form presented by Batchelor [16].

Consider a sphere of radius r_0 and volume V_0. The area of the interface contained within this volume is

$$A_s = \pi r_0^2. \tag{22}$$

The rate at which forces do work on the external boundary A_0 of volume V_0 is

$$W = \int_{A_0}(\mathbf{v}^{(0)}\cdot\boldsymbol{\sigma}\cdot\mathbf{n})dA. \tag{23}$$

In the case of an effective, continuum interface with effective surface shear viscosity η_s^{eff}, this rate equals

$$W = W_0 + 4\eta_s^{\mathrm{eff}}\alpha^2 A_s, \tag{24}$$

where

$$W_0 = \int_{A_0}(\mathbf{v}^{(0)}\cdot\boldsymbol{\sigma}^{(0)}\cdot\mathbf{n})dA. \tag{25}$$

We now proceed to determine the rate in the case when particles are explicitly accounted for. We have

$$W = W_0 + \int_{A_0} (\mathbf{v}^{(0)} \cdot \boldsymbol{\sigma}^{(1)} \cdot \mathbf{n}) dA. \tag{26}$$

The rates in both cases should be equal, therefore

$$4\eta_s^{\text{eff}} \alpha^2 A_s = \int_{A_0} (\mathbf{v}^{(0)} \cdot \boldsymbol{\sigma}^{(1)} \cdot \mathbf{n}) dA. \tag{27}$$

Following Batchelor [16], we transform the surface integral over A_0 to the volume integral over V_0:

$$\begin{aligned} \int_{A_0} (\mathbf{v}^{(0)} \cdot \boldsymbol{\sigma}^{(1)} \cdot \mathbf{n}) dA &= \int_{V_0} ((\boldsymbol{\alpha} \cdot \mathbf{r}) \cdot (\nabla \cdot \boldsymbol{\sigma}^{(1)}) + \boldsymbol{\alpha} : \boldsymbol{\sigma}^{(1)}) dV \\ &\quad - \int_{\sum A_p} [(\boldsymbol{\alpha} \cdot \mathbf{r}) \cdot (\boldsymbol{\sigma}^{(1)} \cdot \mathbf{n})] dA, \end{aligned} \tag{28}$$

where $\sum_{A_p}(\ldots)$ is the sum of the surface areas of the particles contained within volume V_0. Using the equations of motion, Eq. (3), and the relation

$$\begin{aligned} \int_{V_0} \boldsymbol{\alpha} : \boldsymbol{\sigma}^{(1)} dV &= \int_{V_0} 2\eta [\boldsymbol{\alpha} : (\nabla \cdot \mathbf{v}^{(1)})] dV \\ &= - \int_{\sum A_p} 2\eta (\mathbf{v}^{(1)} \cdot \boldsymbol{\alpha} \cdot \mathbf{n}) dA, \end{aligned} \tag{29}$$

valid due to the boundary condition $\mathbf{v}^{(1)} = \mathbf{0}$ at A_0, we obtain

$$\begin{aligned} \int_{A_0} (\mathbf{v}^{(0)} \cdot \boldsymbol{\sigma}^{(1)} \cdot \mathbf{n}) dA &= \sum \int_{A_p} [(\boldsymbol{\alpha} \cdot \mathbf{r}) \cdot (\boldsymbol{\sigma}^{(1)} \cdot \mathbf{n}) - 2\eta (\boldsymbol{\alpha} : \mathbf{v}^{(1)} \mathbf{n})] dA, \end{aligned} \tag{30}$$

which has the same form as the expression given by Batchelor [16], except that the stress tensor now contains interfacial part. The integral of the corresponding contribution over the surface of a particle,

$$\int_{A_p} (\mathbf{v}^{(0)} \cdot \boldsymbol{\sigma}_s \cdot \mathbf{n}) \delta(z) dA, \tag{31}$$

has the form

$$- \alpha R^2 \int_0^{2\pi} \Pi^{(1)} \cos 2\varphi d\varphi. \tag{32}$$

As a result, the equality of the viscous energy dissipation rates in two cases, Eq. (27), becomes

$$\begin{aligned} 4\eta_s^{\text{eff}} \alpha^2 A_s &= -\alpha R^2 N \int_0^{2\pi} \Pi^{(1)}(R) \cos 2\varphi d\varphi \\ &\quad + N \int_{A_p} \left[(\boldsymbol{\alpha} \cdot \mathbf{r}) \cdot \left(\boldsymbol{\sigma}_b^{(1)} \cdot \mathbf{n} \right) - 2\eta (\boldsymbol{\alpha} : \mathbf{v}^{(1)} \mathbf{n}) \right] dA, \end{aligned} \tag{33}$$

yielding the following expression for the effective surface shear viscosity:

$$\begin{aligned} \eta_s^{\text{eff}} &= -\frac{\phi}{4\pi\alpha} \int_0^{2\pi} \Pi^{(1)}(R) \cos 2\varphi d\varphi + \\ &\quad + \frac{\phi}{4\pi R^2 \alpha^2} \int_{A_p} \left[(\boldsymbol{\alpha} \cdot \mathbf{r}) \cdot \left(\boldsymbol{\sigma}_b^{(1)} \cdot \mathbf{n} \right) - 2\eta (\boldsymbol{\alpha} : \mathbf{v}^{(1)} \mathbf{n}) \right] dA. \end{aligned} \tag{34}$$

This expression contains hydrodynamic fields evaluated at the surface of a single particle, half-immersed at an incompressible fluid interface and subjected to unperturbed shear flow given by Eq. (14).

IV. RESULT

In order to obtain the value of the effective surface shear viscosity, we need to calculate the integrals that enter Eq. (34). For this, we need to know the hydrodynamic fields at the surface of the particles. They can be obtained by solution of hydrodynamic equations for a shear flow in the system with a single adsorbed particle.

The details of the numerical solution using vector spherical harmonics representation are given in the Appendix. As a result of calculation, the effective surface shear viscosity is

$$\eta_s^{\text{eff}} = K \eta_{s0}^{\text{eff}}, \tag{35}$$

where

$$\eta_{s0}^{\text{eff}} = \tfrac{5}{3}(\eta_1 + \eta_2) R \phi \tag{36}$$

is the effective surface shear viscosity without account of the interface incompressibility [9], and the coefficient $K = 1.076$.

In the rest of this section the conditions on the particle concentration and shear rate are given under which the result should be valid. They follow from the model assumptions listed in Sec. II.

Concentration of particles has to be small:

$$\phi \ll 1. \tag{37}$$

Lattice Boltzmann modeling of particle-laden flow without account of incompressibility has demonstrated excellent agreement of the effective surface shear viscosity with Eq. (36) for the values of ϕ up to ~ 0.15 [9].

On the other hand, surface shear viscosity of the fluid interface can be neglected if it is small compared to the effective surface shear viscosity due to particles. This gives another condition on the concentration of particles:

$$\phi \gg \frac{\eta_s}{\eta R}. \tag{38}$$

Several model assumptions require shear rate to be small. First, Reynolds number has to be small to allow neglecting inertial effects:

$$\alpha \ll \frac{\eta}{\rho R^2}. \tag{39}$$

Second, surface tension τ has to be large for the interface to be flat:

$$\alpha \ll \frac{\tau}{|\eta_1 - \eta_2| R}. \tag{40}$$

Finally, surface compressibility κ_s has to be small for the interface to be incompressible [10]:

$$\alpha \ll \frac{1}{\kappa_s \eta R}. \tag{41}$$

V. CONCLUDING REMARKS

The effective surface shear viscosity of a flat, low-concentration, particle-laden incompressible interface separating two immiscible fluids has been found to be 7.6% larger than the value obtained without account for surface incompressibility.

Fischer *et al.* [10] calculated the drag on a sphere immersed and moving in an incompressible monolayer. On a monolayer or membrane of low viscosity the translational drag on a half-immersed sphere is 25% larger than the drag on a sphere immersed in a free surface. The present study shows that the effect of a low-viscosity monolayer on the effective viscosity of the particle-laden fluid interface is less than on the drag coefficient of a sphere. Nevertheless, it is not negligibly small and may be detected experimentally.

The present result was obtained in assumption of an infinite system size. The question remains open whether other boundaries located at finite distance, which is the case for real systems, influences the obtained value of the effective surface shear viscosity.

We finally note that the result for the surface dilatational viscosity of low-concentration particle-laden interfaces [9] cannot be extended to the case of viscoelastic fluid interface in the same way because the surface flow in this case is then essentially compressible, which should be explicitly taken into account.

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APPENDIX: HYDRODYNAMIC FIELDS

In this appendix we solve hydrodynamic equations for a single particle at an incompressible interface between two fluids in a shear flow given by Eq. (14). The general approach to the motion of particles in the presence of an incompressible fluid interface was developed by Blawzdziewicz and coworkers Refs. [17–19]. To facilitate the solution, we will make use of the symmetry of the problem and represent hydrodynamic fields in vector spherical harmonics, and their two-dimensional analog, vector polar harmonics.

1. Vector spherical harmonics

Vector spherical harmonics are an extension of the scalar spherical harmonics for use with vector fields. Vector spherical harmonics can be introduced in different ways [20–22]. Vector spherical harmonics representation is convenient for solving differential equations involving vector fields, such as equations of hydrodynamics [23] or electrodynamics [21,24].

We follow Ref. [21] and define vector spherical harmonics as

$$\mathbf{Y}_{lm}(\theta,\varphi) = Y_{lm}\mathbf{e}_r, \tag{A1a}$$

$$\mathbf{\Psi}_{lm}(\theta,\varphi) = r\nabla Y_{lm}, \tag{A1b}$$

$$\mathbf{\Phi}_{lm}(\theta,\varphi) = \mathbf{r}\times\nabla Y_{lm}, \tag{A1c}$$

where $l = 0,\dots,\infty$, $m = -l,\dots,l$, $\mathbf{e}_r$, $\mathbf{e}_\theta$, and $\mathbf{e}_\varphi$ are orts in spherical coordinate system (r, θ, φ), and scalar spherical harmonics $Y_{lm}(\theta,\varphi)$ are defined as

$$Y_{lm}(\theta,\varphi) = (-1)^m\sqrt{\frac{(2l+1}{4\pi}\frac{(l-m)!}{(l+m)!}}P_l^m(\cos\theta)e^{im\varphi}, \tag{A2}$$

where

$$P_l^m(x) = \frac{(-1)^m}{2^l l!}(1-x^2)^{m/2}\frac{d^{l+m}}{dx^{l+m}}(x^2-1)^l \tag{A3}$$

are associated Legendre polynomials [25].

2. Vector polar harmonics

To represent fields at a planar fluid interface we introduce vector polar harmonics in a way analogous to vector spherical harmonics:

$$\mathbf{y}_m(\varphi) = y_m(\varphi)\mathbf{e}_\rho, \tag{A4a}$$

$$\boldsymbol{\psi}_m(\varphi) = r\nabla y_m(\varphi), \tag{A4b}$$

where $m = -\infty,\dots+\infty$, and

$$y_m(\varphi) = \frac{1}{\sqrt{2\pi}}e^{im\varphi} \tag{A5}$$

are scalar polar harmonics.

Vector polar harmonics are related to vector spherical harmonics on a $z = 0$ plane as

$$\mathbf{Y}_{lm}\left(\frac{\pi}{2},\varphi\right) = \chi_{lm}\mathbf{y}_m(\varphi), \tag{A6a}$$

$$\mathbf{\Psi}_{lm}\left(\frac{\pi}{2},\varphi\right) = \chi_{lm}\boldsymbol{\psi}_m(\varphi), \tag{A6b}$$

$$\mathbf{\Phi}_{lm}\left(\frac{\pi}{2},\varphi\right) = -\frac{i}{m}\xi_{lm}\boldsymbol{\psi}_m(\varphi), \tag{A6c}$$

where the quantities χ_{lm} and ξ_{lm} are defined by relations

$$Y_{lm}\left(\frac{\pi}{2},\varphi\right) = \chi_{lm}y_m(\varphi) \tag{A7}$$

and

$$\left.\frac{\partial Y_{lm}(\theta,\varphi)}{\partial\theta}\right|_{\theta=\pi/2} = \xi_{lm}y_m(\varphi). \tag{A8}$$

The quantities χ_{lm} are explicitly given by formula

$$\chi_{lm} = \frac{(-1)^{(l-m)/2}}{2^l}\sqrt{\frac{2l+1}{2}}\frac{\sqrt{(l+m)!(l-m)!}}{\left(\frac{l+m}{2}\right)!\left(\frac{l-m}{2}\right)!}, \tag{A9}$$

for even $l+m$, and are zero otherwise. The quantities ξ_{lm} are explicitly given by formula

$$\xi_{lm} = \frac{(-1)^{(l-m+1)/2}}{2^l}\sqrt{\frac{2l+1}{2}} \times (l+m+1)\frac{\sqrt{(l+m)!(l-m)!}}{\left(\frac{l+m+1}{2}\right)!\left(\frac{l-m-1}{2}\right)!}, \quad \text{(A10)}$$

for odd $l+m$, and are zero otherwise.

3. Representation of hydrodynamic fields

The velocity field $\mathbf{v}^{(1)}(\mathbf{r})$ is expanded in vector spherical harmonics as

$$\mathbf{v}^{(1)}(\mathbf{r}) = \sum_{lm}\left[v_{lm}^{(r)}(r)\mathbf{Y}_{lm}(\theta,\varphi) + v_{lm}^{(1)}(r)\mathbf{\Psi}_{lm}(\theta,\varphi) + v_{lm}^{(2)}(r)\mathbf{\Phi}_{lm}(\theta,\varphi)\right]. \quad \text{(A11)}$$

The pressure field $p^{(1)}(\mathbf{r})$, being scalar, is expanded in scalar spherical harmonics:

$$p^{(1)}(\mathbf{r}) = \sum_{lm} p_{lm}(r)Y_{lm}(\theta,\varphi). \quad \text{(A12)}$$

The surface velocity field $\mathbf{v}_s(\mathbf{r})$ is expanded in vector polar harmonics as

$$\mathbf{v}_s(\mathbf{r}) = \sum_{m=-\infty}^{\infty}\left[u_m^{(\rho)}(r)\mathbf{y}_m(\varphi) + u_m^{(\varphi)}(r)\boldsymbol{\psi}_m(\varphi)\right]. \quad \text{(A13)}$$

The surface pressure field $\Pi^{(1)}(\mathbf{r})$ is expanded in scalar polar harmonics:

$$\Pi^{(1)}(\mathbf{r}) = \sum_{m=-\infty}^{\infty}\Pi_m(r)y_m(\varphi). \quad \text{(A14)}$$

4. Differential operators

This subsection presents explicit expressions for the differential operators used in subsequent derivation.

The expressions for the gradient of the scalar field, the divergence of the vector field, and the curl of the vector field, expanded in vector spherical harmonics, are [21]

$$\nabla p(\mathbf{r}) = \sum_{lm}\left[\frac{\partial p_{lm}(r)}{\partial r}\mathbf{Y}_{lm}(\theta,\varphi) + \frac{p_{lm}(r)}{r}\mathbf{\Psi}_{lm}(\theta,\varphi)\right], \quad \text{(A15)}$$

$$\nabla\cdot\mathbf{v}^{(1)}(\mathbf{r}) = \sum_{lm}\left[\frac{\partial v_{lm}^{(r)}(r)}{\partial r} + \frac{2}{r}v_{lm}^{(r)}(r) - \frac{l(l+1)}{r}v_{lm}^{(1)}(r)\right] \times Y_{lm}(\theta,\varphi), \quad \text{(A16)}$$

$$\nabla\times\mathbf{v}^{(1)}(\mathbf{r}) = \sum_{lm}\Bigg\{-\frac{l(l+1)}{r}v^{(2)}(r)\mathbf{Y}_{lm}(\theta,\varphi) - \left[\frac{\partial v^{(2)}(r)}{\partial r} + \frac{1}{r}v^{(2)}(r)\right]\mathbf{\Psi}_{lm}(\theta,\varphi) + \left[-\frac{1}{r}v_{lm}^{(r)}(r) + \frac{\partial v^{(1)}(r)}{\partial r} + \frac{1}{r}v_{lm}^{(1)}(r)\right]\mathbf{\Phi}_{lm}(\theta,\varphi)\Bigg\}, \quad \text{(A17)}$$

where

$$\sum_{lm}(\cdots) \equiv \sum_{l=0}^{\infty}\sum_{m=-l}^{l}(\cdots). \quad \text{(A18)}$$

Consequently, the Laplacian of a divergence-free vector field is

$$\begin{aligned}\nabla^2\mathbf{v}^{(1)}(\mathbf{r}) &= -\nabla\times[\nabla\times\mathbf{v}^{(1)}(\mathbf{r})]\\ &= \sum_{lm}\Bigg\{-\frac{l(l+1)}{r}\left[\frac{v_{lm}^{(r)}(r)}{r} - \frac{\partial v_{lm}^{(1)}(r)}{\partial r} - \frac{v_{lm}^{(1)}(r)}{r}\right]\mathbf{Y}_{lm}(\theta,\varphi)\\ &\quad - \left[\frac{1}{r}\frac{\partial v_{lm}^{(r)}(r)}{\partial r} - \frac{\partial^2 v_{lm}^{(1)}(r)}{\partial r^2} - \frac{2}{r}\frac{\partial v_{lm}^{(1)}(r)}{\partial r}\right]\mathbf{\Psi}_{lm}(\theta,\varphi)\\ &\quad + \left[\frac{\partial^2 v_{lm}^{(2)}(r)}{\partial r^2} + \frac{2}{r}\frac{\partial v_{lm}^{(2)}(r)}{\partial r} - \frac{l(l+1)}{r^2}v_{lm}^{(2)}(r)\right]\mathbf{\Phi}_{lm}(\theta,\varphi)\Bigg\}.\end{aligned} \quad \text{(A19)}$$

The surface gradient of a scalar field and the surface divergence of a vector field are expanded in polar harmonics as

$$\nabla_s\Pi^{(1)}(\mathbf{r}) = \sum_{m=-\infty}^{\infty}\left[\frac{\partial\Pi_m(r)}{\partial r}\mathbf{y}_{lm}(\varphi) + \frac{\Pi_m(r)}{r}\boldsymbol{\psi}_{lm}(\varphi)\right], \quad \text{(A20)}$$

$$\nabla_s\cdot\mathbf{v}_s^{(1)}(\mathbf{r}) = \sum_{m=-\infty}^{\infty}\left[\frac{\partial v_m^{(\rho)}(r)}{\partial r} + \frac{1}{r}v_m^{(\rho)}(r) - \frac{m^2}{r}v_m^{(\varphi)}(r)\right]y_m. \quad \text{(A21)}$$

5. Boundary conditions

The boundary conditions at infinity $\mathbf{v}^{(1)}(\infty)=0$ and $p^{(1)}(\infty)=0$ can be represented as

$$v_{lm}^{(r)}(\infty) = 0, \quad \text{(A22a)}$$

$$v_{lm}^{(1)}(\infty) = 0, \quad \text{(A22b)}$$

$$v_{lm}^{(2)}(\infty) = 0, \quad \text{(A22c)}$$

$$p_{lm}(\infty) = 0. \quad \text{(A22d)}$$

At particle surface we have $\mathbf{v}(R)=0$, which corresponds to $\mathbf{v}^{(1)}(R) = -\mathbf{v}^{(0)}(R)$, or

$$v_x^{(1)}(R) = -\alpha x, \quad \text{(A23a)}$$

$$v_y^{(1)}(R) = \alpha y, \quad \text{(A23b)}$$

$$v_z^{(1)}(R) = 0. \quad \text{(A23c)}$$

These conditions are equivalent to

$$u_{2,\pm 2}^{(r)}(R) = -2\sqrt{\frac{2\pi}{15}}\alpha R, \quad \text{(A24a)}$$

$$u_{2,\pm 2}^{(1)}(R) = -\sqrt{\frac{2\pi}{15}}\alpha R, \quad \text{(A24b)}$$

$$u_{2,\pm 2}^{(2)}(R) = 0, \quad \text{(A24c)}$$

$$u_{l,m}^{(r,1,2)}(R) = 0 \quad \text{(for other values of } l \text{ and } m). \quad \text{(A24d)}$$

6. Surface continuity equation

By means of Eq. (A21) for surface divergence of a velocity field, surface incompressibility condition Eq. (10) is equivalent to the relations

$$\frac{\partial v_m^{(\rho)}(r)}{\partial r} + \frac{1}{r} v_m^{(\rho)}(r) - \frac{m^2}{r} v_m^{(\varphi)}(r) = 0, \quad m = -\infty, \dots, +\infty. \tag{A25}$$

Writing the surface velocity as

$$\mathbf{v}_s(\mathbf{r}) = \sum_{lm} \left\{ \chi_{lm} v_{lm}^{(r)}(r) \mathbf{y}_m(\varphi) + \left[\chi_{lm} v_{lm}^{(1)}(r) - \frac{i}{m} \xi_{lm} v_{lm}^{(2)}(r) \right] \boldsymbol{\psi}_m(\varphi) \right\}, \tag{A26}$$

we can represent surface divergence of a velocity field as

$$\nabla_s \cdot \mathbf{v}_s(\mathbf{r}) = \sum_{lm} \left\{ \chi_{lm} \left[\frac{\partial v_{lm}^{(r)}(r)}{\partial r} + \frac{1}{r} v_{lm}^{(r)}(r) - \frac{m^2}{r} v_{lm}^{(1)}(r) \right] + \xi_{lm} \frac{im}{r} v_{lm}^{(2)}(r) \right\} y_m(\varphi). \tag{A27}$$

The boundary conditions at the fluid interface thus become

$$\sum_{l=|m|}^{\infty} \left\{ \chi_{lm} \left[\frac{\partial v_{lm}^{(r)}(r)}{\partial r} + \frac{1}{r} v_{lm}^{(r)}(r) - \frac{m^2}{r} v_{lm}^{(1)}(r) \right] + \xi_{lm} \frac{im}{r} v_{lm}^{(2)}(r) \right\} = 0. \tag{A28}$$

7. Bulk continuity equation

Since unperturbed flow satisfies the equation $\nabla \cdot \mathbf{v}^{(0)}(\mathbf{r}) = 0$, bulk continuity Eq. (7) yields

$$\nabla \cdot \mathbf{v}^{(1)}(\mathbf{r}) = 0. \tag{A29}$$

Using Eq. (A16) for the divergence of the vector field expanded in vector spherical harmonics, the continuity equation can be represented as

$$\frac{\partial v_{lm}^{(r)}(r)}{\partial r} + \frac{2}{r} v_{lm}^{(r)}(r) = \frac{l(l+1)}{r} v_{lm}^{(1)}(r), \quad l = 0, \dots, \infty, \quad m = -l, \dots, l. \tag{A30}$$

8. Stokes equations

The equations of motion, Eq. (3), with the stress tensor given by Eq. (4),

$$\eta \nabla^2 \mathbf{v}(\mathbf{r}) = \nabla p(\mathbf{r}) + \delta(z) \nabla_s \Pi(\mathbf{r}), \tag{A31}$$

yield the equations for the perturbation fields only:

$$\eta \nabla^2 \mathbf{v}^{(1)}(\mathbf{r}) = \nabla p^{(1)}(\mathbf{r}) + \delta(z) \nabla_s \Pi^{(1)}(\mathbf{r}). \tag{A32}$$

We will write these equations in vector spherical harmonic representation.

We write the following expansions of the surface (Marangoni) force,

$$\mathbf{f}(\mathbf{r}) = -\nabla \Pi^{(1)}(\mathbf{r}), \tag{A33}$$

into vector (polar and spherical) harmonics:

$$\mathbf{f}(\mathbf{r}) = \sum_{m=-\infty}^{\infty} \left[f_m^{(\rho)}(r) \mathbf{y}_m(\varphi) + f_m^{(\varphi)}(r) \boldsymbol{\psi}_m(\varphi) \right], \tag{A34}$$

$$\delta(z) \mathbf{f}(\mathbf{r}) = \sum_{lm} \left[f_{lm}^{(r)}(r) \mathbf{Y}_{lm}(\theta,\varphi) + f_{lm}^{(1)}(r) \boldsymbol{\Psi}_{lm}(\theta,\varphi) + f_{lm}^{(2)}(r) \boldsymbol{\Phi}_{lm}(\theta,\varphi) \right]. \tag{A35}$$

Taking into account that

$$\int_0^{2\pi} \mathbf{y}_m(\varphi) \cdot \mathbf{Y}_{lm}^* \left(\frac{\pi}{2}, \varphi \right) d\varphi = \chi_{lm}, \tag{A36a}$$

$$\int_0^{2\pi} \boldsymbol{\psi}_m(\varphi) \cdot \boldsymbol{\Psi}_{lm}^* \left(\frac{\pi}{2}, \varphi \right) d\varphi = m^2 \chi_{lm}, \tag{A36b}$$

$$\int_0^{2\pi} \boldsymbol{\psi}_m(\varphi) \cdot \boldsymbol{\Phi}_{lm}^* \left(\frac{\pi}{2}, \varphi \right) d\varphi = im\xi_{lm}, \tag{A36c}$$

we obtain the following relations:

$$f_{lm}^{(r)}(r) = \frac{\chi_{lm}}{r} f_m^{(\rho)}(r), \tag{A37a}$$

$$f_{lm}^{(1)}(r) = \frac{m^2 \chi_{lm}}{l(l+1)r} f_m^{(\varphi)}(r), \tag{A37b}$$

$$f_{lm}^{(2)}(r) = \frac{im\xi_{lm}}{l(l+1)r} f_m^{(\varphi)}(r). \tag{A37c}$$

Substituting Eq. (A14) in Eq. (A33), and using Eq. (A20), we can write

$$f_{lm}^{(r)}(r) = -\frac{\chi_{lm}}{r} \frac{\partial \Pi_m(r)}{\partial r}, \tag{A38a}$$

$$f_{lm}^{(1)}(r) = -\frac{m^2 \chi_{lm}}{l(l+1)r^2} \Pi_m(r), \tag{A38b}$$

$$f_{lm}^{(2)}(r) = -\frac{im\xi_{lm}}{l(l+1)r^2} \Pi_m(r). \tag{A38c}$$

Combining the above expressions, we obtain the following form of Stokes equations:

$$-\eta \frac{l(l+1)}{r} \left[\frac{1}{r} v_{lm}^{(r)}(r) - \frac{\partial v_{lm}^{(1)}(r)}{\partial r} - \frac{1}{r} v_{lm}^{(1)}(r) \right] = \frac{\partial p_{lm}(r)}{\partial r} + \frac{\chi_{lm}}{r} \frac{\partial \Pi_m(r)}{\partial r}, \tag{A39a}$$

$$-\eta \left[\frac{1}{r} \frac{\partial v_{lm}^{(r)}(r)}{\partial r} - \frac{\partial^2 v_{lm}^{(1)}(r)}{\partial r^2} - \frac{2}{r} \frac{\partial v_{lm}^{(1)}(r)}{\partial r} \right] = \frac{1}{r} p_{lm}(r) + \frac{m^2 \chi_{lm}}{l(l+1)r^2} \Pi_m(r), \tag{A39b}$$

$$\eta \left[\frac{\partial^2 v_{lm}^{(2)}(r)}{\partial r^2} + \frac{2}{r} \frac{\partial v_{lm}^{(2)}(r)}{\partial r} - \frac{l(l+1)}{r^2} v_{lm}^{(2)}(r) \right] = \frac{im\xi_{lm}}{l(l+1)r^2} \Pi_m(r). \tag{A39c}$$

9. Solution of hydrodynamic equations

We are solving the Stokes equations, Eqs. (A30) and (A39), and surface continuity equation, Eq. (A28), with boundary condition Eqs. (A22) and (A24).

By introducing functions $P_{lm}(r)$ satisfying $p_{lm}(r) = dP_{lm}(r)/dr$, we can write these equations in form

$$\frac{dv^{(r)}_{lm}(r)}{dr} + \frac{2}{r}v^{(r)}_{lm}(r) - \frac{l(l+1)}{r}v^{(1)}_{lm}(r) = 0, \tag{A40a}$$

$$\eta\frac{l(l+1)}{r}\left[\frac{1}{r}v^{(r)}_{lm}(r) - \frac{dv^{(1)}_{lm}(r)}{dr} - \frac{1}{r}v^{(1)}_{lm}(r)\right] + \frac{d^2P_{lm}(r)}{dr^2} + \frac{\chi_{lm}}{r}\frac{d\Pi_m(r)}{dr} = 0, \tag{A40b}$$

$$\eta\left[\frac{1}{r}\frac{dv^{(r)}_{lm}(r)}{dr} - \frac{d^2v^{(1)}_{lm}(r)}{dr^2} - \frac{2}{r}\frac{dv^{(1)}_{lm}(r)}{dr}\right] + \frac{1}{r}\frac{dP_{lm}(r)}{dr} + \frac{m^2\chi_{lm}}{l(l+1)r^2}\Pi_m(r) = 0, \tag{A40c}$$

$$\eta\left[\frac{d^2v^{(2)}_{lm}(r)}{dr^2} + \frac{2}{r}\frac{dv^{(2)}_{lm}(r)}{dr} - \frac{l(l+1)}{r^2}v^{(2)}_{lm}(r)\right] - \frac{im\xi_{lm}}{l(l+1)r^2}\Pi_m(r) = 0, \tag{A40d}$$

$$\sum_{l=|m|}^{\infty}\left\{\chi_{lm}\left[\frac{dv^{(r)}_{lm}(r)}{dr} + \frac{1}{r}v^{(r)}_{lm}(r) - \frac{m^2}{r}v^{(1)}_{lm}(r)\right] + \xi_{lm}\frac{im}{r}v^{(2)}_{lm}(r)\right\} = 0, \tag{A40e}$$

where $l = 2,\ldots,\infty$, and $m = \pm 2$ due to boundary conditions at the surface of the particle [Eqs. (A24)]. This is an infinite system of Cauchy-Euler equations for functions $v^{(r)}_{lm}(r)$, $v^{(1)}_{lm}(r)$, $v^{(2)}_{lm}(r)$, $P_{lm}(r)$, and $\Pi_m(r)$. By means of substitution $r = e^t$ it is equivalent to an infinite system of linear homogeneous differential equations with constant coefficients. The differential operator of this system can be shown to be bounded [26]; therefore, we can employ the method of reduction [27] and truncate the infinite system Eqs. (A40) at some value $l = l_{\max}$. The solution of the infinite system will then be given by a limit of the solution of the finite system at $l_{\max} \to \infty$.

As a result, the solution of the system Eqs. (A40) with boundary condition Eqs. (A22) and (A24) has the form

$$\Pi_m(r) = \sum_n \frac{c_{m,n}}{r^n}, \tag{A41a}$$

$$v^{(r)}_{lm}(r) = \sum_n \frac{c^{(r)}_{l,m,n}}{r^n}, \tag{A41b}$$

$$v^{(1)}_{lm}(r) = \sum_n \frac{c^{(1)}_{l,m,n}}{r^n}, \tag{A41c}$$

$$v^{(2)}_{lm}(r) = \sum_n \frac{c^{(2)}_{l,m,n}}{r^n}, \tag{A41d}$$

$$p_{lm}(r) = \sum_n \frac{c^{(p)}_{l,m,n}}{r^{n+1}}, \tag{A41e}$$

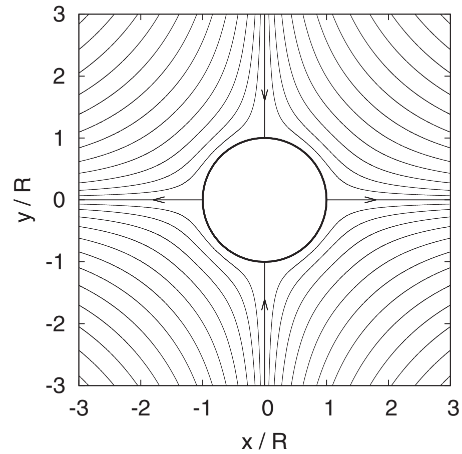

FIG. 1. Streamlines of the interfacial flow ($z = 0$) near the particle.

where

$$n = 2,3,5,7,9,\ldots, \tag{A42}$$

and the the values of the coefficients $c_{m,n}$, $c^{(r)}_{l,m,n}$, $c^{(1)}_{l,m,n}$, $c^{(2)}_{l,m,n}$, and $c^{(p)}_{l,m,n}$ are provided by numerical calculation.

This solution corresponds to a shear flow around a particle adsorbed at the incompressible fluid interface (see Fig. 1). Velocity at large distance decays as $1/r^2$, but the prefactor is greater due to incompressibility of the interfacial flow (see Fig. 2).

10. Calculation of surface viscosity

In order to obtain the value of the effective surface shear viscosity, we need to calculate the integrals that enter Eq. (34). Substituting vector harmonic expansions of hydrodynamic

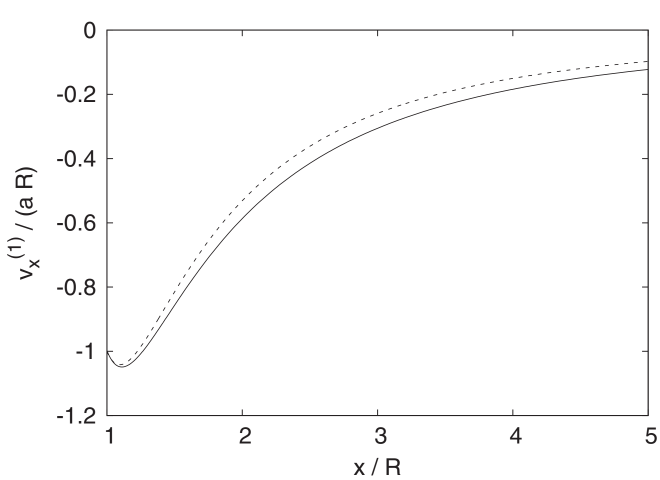

FIG. 2. Perturbation velocity component $v^{(1)}_x$ at $y = 0$, $z = 0$ as a function of the distance x from the center of the particle with (solid line) and without (dashed line) account for incompressibility of interfacial flow.

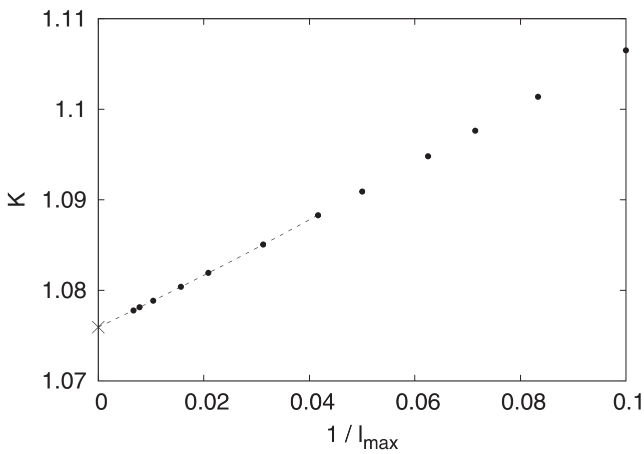

FIG. 3. The coefficient K, defined by Eq. (35), as a function of the truncation parameter $l_{\max}$. Dashed line corresponds to quadratic extrapolation, cross indicates the value of K at $l_{\max} \to \infty$.

fields given in Appendix A3, we obtain

$$\int_0^{2\pi} \Pi^{(1)} \cos 2\varphi d\varphi = \sqrt{2\pi}\,\Pi_2(R), \tag{A43}$$

$$\int_{A_p} [(\boldsymbol{\alpha}\cdot\mathbf{r})\cdot(\boldsymbol{\sigma}^{(1b)}\cdot\mathbf{n}) - 2\eta(\boldsymbol{\alpha}:\mathbf{v}^{(1)}\mathbf{n})]dA = \sqrt{\frac{32\pi}{15}}\big\{v_{2,2}^{(r)}(R) + 2\big[v_{2,2}^{(r)}\big]'(R) - 9v_{2,2}^{(1)}(R) + 3\big[v_{2,2}^{(1)}\big]'(R) - p_{2,2}(R)\big\}, \tag{A44}$$

where the prime indicates differentiation with respect to r, and the units are chosen such that $\eta = 1$, $\alpha = 1$, $R = 1$.

Using the numerical solution described in Appendix A9, we can calculate the values of the expression Eqs. (A43) and (A44) and, consequently, the coefficient K, defined by Eq. (35), as a function of the truncation parameter $l_{\max}$ (see Fig. 3). Numerical calculation of the limit $l_{\max} \to \infty$ yields $K = 1.076$.

11. Viscosity contrast

The above derivation used the assumption that the shear viscosities of both bulk fluids are equal. In the case of different viscosities the hydrodynamic fields still satisfy the same equations and boundary conditions. Therefore, the velocity field remains the same. Viscous dissipation in the domain occupied by each bulk fluid will be proportional to the value of corresponding shear viscosity; therefore, the result given by Eq. (35) remains valid in the case of velocity contrast.

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