Hydrodynamic coupling of spherical particles to a planar fluid-fluid interface: Theoretical analysis

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We have developed a new technique (based on our Cartesian-representation method) to describe hydrodynamic interactions of a spherical particle with an undeformable planar fluid-fluid interface under creeping-flow conditions. The interface can be either surfactant-free or covered with an incompressible surfactant monolayer. We consider the effect of surface incompressibility and surface viscosity on particle motion. The new algorithm allows to calculate particle mobility coefficients for hydrodynamically coupled particles, moving either on the same or on the opposite sides of the interface. © 2010 American Institute of Physics. [doi:10.1063/1.3475217]

I. INTRODUCTION

Hydrodynamic interactions of suspended particles with planar fluid interfaces,¹,² membranes,³ and rigid walls⁴⁻¹¹ are important in soft-condensed-matter physics. Applications in which such interactions play a significant role include measuring interfacial transport coefficients (e.g., interfacial viscosity),¹² probing nonhydrodynamic forces by observing near-wall particle motion,¹³⁻¹⁵ evaluating hydrodynamic hindrance corrections in near-wall particle-imaging velocimetry,¹⁶ and estimating collection efficiency for large drops moving in a suspension of fine particles.¹⁷,¹⁸ Particle-interface hydrodynamic interactions also affect dynamics of particle-stabilized thin liquid films,¹⁹ enhance hydrodynamic diffusivity near interfaces,²⁰,²¹ and result in complex collective particle behavior in parallel-wall channels.²²⁻²⁴

The motion of spherical particles immersed in a fluid bounded by a planar rigid wall has been studied in much detail.²⁻⁵,¹⁹,⁲²⁻⁵² However, available results for fluid-fluid interfaces with different properties are much more limited. For example, most of the previous investigations have been restricted to the motion of a single sphere¹,¹²,²⁸⁻³¹ and, to our knowledge, hydrodynamic coupling between particles moving on the opposite sides of the interface has not been investigated. Moreover, even for a single particle in a creeping flow regime, the understanding of the effect of interfacial properties on particle dynamics is incomplete (in spite of a significant effort²⁻¹²,³⁰⁻⁳⁵).

We examine the hydrodynamic coupling of rigid spherical particles to a planar interface that separates two immiscible fluids of different viscosities. The interface can be either surfactant-free or covered by a surfactant monolayer. We consider the effect of surfactant incompressibility and surface viscosity on the particle motion. Our analysis of the influence of interfacial properties on the particle dynamics is applicable not only to surfactant-covered interfaces, but also to particles moving near fluid membranes (such as, for example, a lipid bilayer).

To analyze particle interactions with fluid-fluid interfaces of different properties, we adapt concepts from our Cartesian-representation method that describes particle motion near planar rigid walls.⁷,⁸,³⁴ We derive new reflection and transmission matrices representing the interaction of Stokes flow with a fluid-fluid interface and incorporate them into our Cartesian-representation calculation scheme. We also propose a novel method for evaluation of hydrodynamically coupled particles placed on the opposite sides of the interface.

In the accompanying study,³⁵ our new technique is applied to calculate the hydrodynamic friction and mobility coefficients for a single sphere moving near an inviscid (compressible and incompressible) interface. This is the first application of our technique. Our new method can be also used to obtain numerical results for interfaces with nonzero surface viscosity and for many particles moving at opposite sides of the interface. We will return to these problems in forthcoming publications.

Our paper is organized as follows. In Sec. II, we define the system and describe a class of interfaces for which our Cartesian-representation method can be used to evaluate particle-interface hydrodynamic interactions. (This broad class includes fully compressible interfaces and surfactant-covered incompressible interfaces, inviscid or with nonzero interfacial viscosity.) Section III provides a general description of our method. Details of our analysis are presented in Secs. IV–VI, and the concluding remarks are given in Sec. VII.

In Sec. IV we describe interactions between a single particle and an interface, and in Sec. V we show that our method can be used to analyze hydrodynamic coupling of two (or many) particles at the opposite sides of the interface. In Sec. VI we provide a derivation of analytical expressions for reflection and transmission matrices that are central to our

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analysis. Although in this paper we focus on two systems: (a) a single spherical particle moving near a fluid-fluid interface and (b) two hydrodynamically coupled spheres moving at the opposite sides of the interface, our results can readily be generalized to an arbitrary number of particles.

II. PARTICLE IN THE PRESENCE OF PLANAR INTERFACE

A. The system

We consider hydrodynamic interactions of solid spherical particles with a planar undeformable interface separating two immiscible fluids of viscosities $\eta_1$ and $\eta_2$. The interface is in the plane $z=0$, fluid 1 occupies the region $z>0$, and fluid 2 occupies the region $z<0$.

Our goal is to determine the friction matrix $\xi_{ij}$ that relates the force $F_i$ and torque $T_i$ acting on particle $i$ to the linear and angular velocities $U_j$ and $\Omega_j$ of all particles in the system

$$
\begin{bmatrix}
F_i \\
T_i
\end{bmatrix} = \sum_{j=1}^{N} \xi_{ij} \cdot \begin{bmatrix}
U_j \\
\Omega_j
\end{bmatrix}.
$$

We discuss systems with a single solid sphere ($N=1$) and two solid spheres ($N=2$) on the opposite sides of the interface (cf., Fig. 1). Our results, however, are applicable to many-particle systems as well.

We assume that the interface is maintained in its planar equilibrium shape either by sufficiently strong interfacial forces or by gravity. The capillary stabilizing mechanism requires that

$$
Ca = \frac{\eta U}{\sigma^{eq}} \ll 1,
$$

where $Ca$ is the capillary number, $U$ is the particle velocity, and $\sigma^{eq}$ the equilibrium interfacial tension. The gravitational stabilizing mechanism requires that

$$
\frac{\eta U}{gd^2 \Delta \rho} \ll 1,
$$

where $g$ denotes gravitational acceleration and $\Delta \rho$ is the mass density difference of the two fluids. The estimates (2) and (3) indicate that the interface remains undeformed if the particle velocity is sufficiently small (which is a frequently encountered limit).

In our calculations creeping-flow conditions are assumed. The flow of the bulk fluids is described by Stokes equations

$$
\eta_i \nabla^2 u_i - \nabla p_i = 0,
$$

$$
\nabla \cdot u_i = 0.
$$

Here, $u_i$ and $p_i$ are the velocity and pressure fields in a fluid phase $i$. The fluid velocity satisfies the no-slip boundary conditions on the particle surface. The velocity is continuous across the interface and the normal velocity component vanishes

$$
u_1 = u_2 = u_s, \quad \hat{e}_z \cdot u_s = 0, \quad \text{for} \quad z = 0,
$$

where $\hat{e}_z$ is the unit vector normal to the interface, and $u_s$ is the local two-dimensional interfacial velocity.

In Sec. II B we discuss additional boundary conditions adequate for four classes of interfaces: fully compressible and incompressible interfaces that are inviscid or have non-zero interfacial viscosity.

B. Surfactant-free and surfactant-covered interface in Stokes flow

1. Stress boundary conditions

For a clean interface (i.e., free of adsorbed species, such as surfactants), the jump

$$
\delta \tau_i = I_s \cdot (\tau_2 - \tau_1) \cdot \hat{e}_i|_{z=0}
$$

of tangential viscous tractions across the interface $z=0$ vanishes. Here $\tau_1$ is the stress tensor in phase $k=1,2$ and $I_s = \hat{e}_s \hat{e}_s + \hat{e}_z \hat{e}_z$ denotes the projection tensor onto the interface. More generally, the jump of tangential viscous tractions is related to the surface divergence of the interfacial stresses $\tau_i$

$$
\delta \tau_i = \nabla_s \cdot \tau_i,
$$

where $\nabla_s = I_s \cdot \nabla$ is the gradient operator along the interface. For a surfactant covered interface, the surface stress tensor is a combination of the interfacial-tension component and viscous component

$$
\nabla_s \cdot \tau_i = \nabla_s \sigma + \nabla_s \cdot \tau_i^{\text{visc}},
$$

where

$$
\sigma = \sigma(c)
$$

is the interfacial tension that depends on the local surfactant concentration $c$. Similar relations also hold for a fluidlike two-dimensional fluid membrane (e.g., lipid bilayer).

Assuming Newtonian interfacial rheology, the interfacial viscous stress is given by the linear constitutive relation

$$(\hat{e}_z \cdot \tau_i^{\text{visc}}) = \eta \mathbf{e}_z \frac{d \mathbf{e}_z}{dt}.$$
\( \mathbf{\Sigma}^{\text{visc}} = 2\beta \{ \nabla \mathbf{u}_s \}_{ij} + \kappa \nabla_{i} \mathbf{u}_s I_{i}, \)  

(10)

where

\( [\nabla_{i} \mathbf{u}_s]_{ij} = \frac{1}{2} [\nabla_{i} \mathbf{u}_s + (\nabla_{j} \mathbf{u}_s)^{T} - \nabla_{i} \mathbf{u}_s I_{j}] \)

(11)
is the deviatoric part of the surface strain-rate tensor \( \nabla_{i} \mathbf{u}_s \) (with 
\( \text{dagger denoting the transpose} \) and \( \beta \) and \( \kappa \) are 
the surface shear and expansion viscosity coefficients. Equations 
(10) and (11) yield

\[ \nabla_s \cdot \mathbf{\Sigma}^{\text{visc}} = \beta \nabla_{j} \mathbf{u}_s + \kappa \nabla_{i} (\nabla_{i} \cdot \mathbf{u}_s). \]

(12)

**2. Surfactant redistribution**

In general, the Stokes-flow problem (4)–(12) need to be solved along with the surfactant-transport equation

\[ \frac{\partial c}{\partial t} = - \nabla_s \cdot (c \mathbf{u}_s), \]

(13)

which is needed to determine the surface-tension contribution in the stress-balance Eq. (7). The resulting evolution equations are nonlinear and the hydrodynamic force acting on a moving particle depends not only on the instantaneous particle velocity but also on the history of motion.

In the present paper we consider two important limiting cases where the evolution of the surfactant distribution on the interface does not need to be explicitly followed. These limiting cases include (i) a fully compressible interface (where there are no interfacial-tension gradients) and (ii) an incompressible interface (where there is no variation of surfactant concentration). In both cases, the boundary conditions on the interface are linear, which implies that the particle motion can be represented in terms of linear friction relation (1).

**3. Compressible and incompressible interfaces**

\textit{a. Fully compressible interface.} An interface can be treated as fully compressible if interfacial-tension gradients (Marangoni stresses) vanish

\[ \nabla_s \sigma = 0, \]

(14)
or if they can be neglected compared to the viscous stresses acting on the interface. According to Eqs. (7), (8), and (10), we thus have

\[ \mathbf{\delta}_{s} = 0 \]

(15)

for an inviscid interface and

\[ \mathbf{\delta}_{s} = \beta \nabla_{j} \mathbf{u}_s + \kappa \nabla_{i} (\nabla_{i} \cdot \mathbf{u}_s) \]

(16)

for an interface with nonzero surface viscosity.

Relation (14) is valid for clean interfaces because the interfacial tension has no spatial variation (assuming isothermal conditions). In the absence of adsorbed species, not only the Marangoni stress vanishes, but also the interfacial viscosity; therefore the boundary condition (15) applies. In the case of surfactant-covered interfaces, approximation (14) is valid when the changes of the interfacial tension \( \Delta \sigma_{\text{max}} \) due to \( O(1) \) surfactant redistribution are too small to generate a significant Marangoni stress \( \tau_{\text{sr}} \sim \Delta \sigma_{\text{max}}/l_c \) in comparison to the characteristic viscous stress \( \tau_{\text{v}} \sim U \eta_{\text{visc}}/l_c \). Here, \( l_c \) is the characteristic length scale, which is the particle-wall distance \( z \) for our problem.\(^{22}\) Comparing the magnitudes of the interfacial and viscous stresses yields

\[ \Delta \sigma_{\text{max}} \ll \eta_{\text{visc}} U. \]

(17)

Approximation (14) is also valid for high-frequency small-amplitude oscillations of a particle because such oscillations do not produce a significant surfactant redistribution. At high surfactant concentrations, surfactant-covered interfaces often have nonzero surface viscosity.

\textit{b. Incompressible interface.} In the incompressible-surfactant limit, an infinitesimal surfactant redistribution on the interface \( \delta \sigma \) produces interfacial-tension gradients sufficiently large to balance the viscous stresses in the bulk fluids. Therefore, an essentially uniform surfactant concentration is maintained on the interface by the gradient of the interfacial stress (8).

Under these assumptions, the interfacial surfactant-transport Eq. (13) yields the incompressibility condition

\[ \nabla_s \cdot \mathbf{u}_s = 0 \]

(18)

for the interfacial velocity \( \mathbf{u}_s \). In the incompressible-surfactant limit, the surfactant concentration is thus eliminated from the governing equations and the interfacial tension becomes the corresponding independent Lagrange-multiplier field. The gradient of this field

\[ \nabla_s \sigma \neq 0, \]

(19)

assumes the form that is needed to ensure that the interfacial-incompressibility constraint (18) is satisfied. Thus, for incompressible interfacial flows, the interfacial tension plays a similar role to the pressure field for bulk incompressible flows.

In the case of an inviscid incompressible interface the stress-balance relations (7) and (8) yield

\[ \mathbf{\mathbf{\delta}}_{s} = \nabla_s \sigma, \]

(20)

and for an interface with nonzero surface viscosity, relations (7), (8), and (10) yield

\[ \mathbf{\mathbf{\delta}}_{s} = \nabla_s \sigma + \beta \nabla_{j} \mathbf{u}_s. \]

(21)

Together with the surface incompressibility condition (18), the stress-balance condition (20) or (21) forms a closed set of boundary conditions for the tangential velocity and tangential viscous tractions. We note that in Eq. (21) there is no compressional-viscosity contribution because the flow is surface-divergence free.

A surfactant-covered interface is incompressible when

\[ \Delta \sigma_{\text{max}} \gg \eta_{s} U. \]

(22)

Under this condition, typical perturbations of the interfacial tension from equilibrium

\[ \delta \sigma \sim \eta_{s} U \ll \Delta \sigma_{\text{max}} \]

(23)
correspond to small perturbations of the surfactant distribution.
\[ \delta c / c_0 \ll 1 \]  

where \( c_0 \) is the equilibrium surfactant concentration.

The incompressible surfactant conditions are common. For example, taking \( \Delta \sigma_{\text{max}} \approx 10 \ \text{dyn cm}^{-1}, \ \eta_t \approx 10^{-2} \ \text{dyn s cm}^{-2}, \) and \( U \approx 1 \ \text{cm s}^{-1} \) for a particle moving near a water-air interface, we find \( \delta c / c_0 \approx 10^{-3} \), which is within the incompressible-surfactant regime.

As argued in our previous publications, the incompressibility condition (24) is usually fulfilled in the small-capillary-number regime. This is because both the interface deformation and surfactant redistribution are controlled by the magnitude of the viscous stresses produced by the moving particle. Specifically, relations (2) and (23) yield \( \delta \sigma \ll \sigma_0 \), which implies that \( \Delta \sigma \ll \Delta \sigma_{\text{max}} \) unless \( \Delta \sigma_{\text{max}} \ll \sigma_0 \). Thus \( O(1) \) surfactant redistribution can occur only for a dilute surfactant film and for systems close to interfacial phase-transition points, where the surfactant elasticity is small.

The above argument can be reversed: the surfactant incompressibility condition (24) implies that the capillary number \( C_a \) is small. Incompressible interfaces are thus typically also undeformable. Incompressible viscous fluid membranes (e.g., a lipid bilayer) are described by a similar set of boundary conditions.

### III. EVALUATION OF HYDRODYNAMIC INTERACTIONS—AN OVERVIEW

Our Cartesian-representation method combines the multipolar expansion of Stokes flow into spherical harmonics with the Cartesian representation that uses lateral Fourier modes in planes parallel to the interface. Such a dual expansion into two sets of basic fields takes into account the spherical shape of the particle and the planar shape of the interface.

So far our method has been used to study hydrodynamic interactions of solid spheres with rigid walls.\(^{21,23,24,39,40}\) In the present paper we show that this method can be expanded and generalized to study fluid-fluid interfaces with different physical properties (e.g., compressible and incompressible interfaces, and interfaces with a nonzero interfacial viscosity). Moreover, our technique can also account for hydrodynamic interactions of particles placed on the opposite sides of the interface.

In Sec. IV we explain our technique, using as an example a single-particle system. The hydrodynamic problem is formulated in Sec. IV A, where the fluid velocity is decomposed into three components: the flow scattered by a moving particle, the flow reflected by the interface, and the flow transmitted by the interface. These three flow components are related by respective operators describing the particle-scattering, interface-reflection, and interface-transmission.

In Sec. IV B the problem is reformulated in the matrix form by expanding the component flows into spherical and Cartesian sets of basis fields. In our technique, the interaction of the flow with the interface is described via the reflection and transmission matrices. The transmission matrix is a new element introduced into our analysis—there is no transmitted flow for rigid walls that were studied in our previous papers.

By partial inversion of the matrix equations for the expansion coefficients, we derive linear equations for multipolar amplitudes of the flow field produced by the particle moving in the presence of the interface (cf., Sec. IV C). The friction matrix \( \zeta \) can be obtained from these amplitudes using formulas introduced in our previous publications.\(^{7,8}\)

In Sec. V our one-particle calculation scheme is generalized to a system consisting of two particles moving on the opposite sides of the interface. In our analysis we follow steps similar to those described in Sec. IV, but we focus on the role of the flow transmitted by the interface. The results of Sec. V do not have a direct analogy in a system bounded by a rigid wall.

The expressions provided in Secs. IV and V are applicable to interfaces that can be characterized via linear reflection and transmission matrices. Explicit expressions for these matrices are derived in Sec. VI. These expressions are given for fully compressible and incompressible interfaces, with or without surface viscosity. The results obtained in Secs. IV and V, combined with the expressions for reflection and transmission matrices in Sec. VI, yield a very accurate and efficient scheme for evaluating the friction matrix for spherical particles that are hydrodynamically coupled to the interface.

### IV. SINGLE-PARTICLE PROBLEM

#### A. Particle-scattered and wall-reflected flows

As illustrated in Fig. 1(a), we consider a spherical particle of diameter \( d \) translating and rotating with linear and angular velocities \( \mathbf{U} \) and \( \mathbf{\Omega} \) in a viscous fluid bounded by a planar interface separating it from another fluid. Our goal is to determine the torque \( \mathbf{T} \) and force \( \mathbf{F} \) acting on the particle for a given particle motion \( \mathbf{U} \) and \( \mathbf{\Omega} \). In what follows the position of a fluid element will be denoted by \( \mathbf{r}' = (x', y', z') \). We assume that the fluid–fluid interface is at the position \( z = 0 \), and the particle center is at \( \mathbf{r} = (0, 0, z) \) with \( z > 0 \).

**a. Flow decomposition.** To evaluate the fluid flow produced by the moving particle, the velocity field \( \mathbf{u} \) is represented as a combination of the flow scattered by the particle, \( \mathbf{u}^p \), the flow reflected by the interface \( \mathbf{u}^R \) (for \( z' \geq 0 \)), and the flow transmitted by the interface \( \mathbf{u}^T \) (for \( z' \leq 0 \)).

\[
\mathbf{u} = \mathbf{u}^p + \mathbf{u}^R, \quad z' \geq 0, \quad (25a)
\]

\[
\mathbf{u} = \mathbf{u}^T, \quad z' \leq 0. \quad (25b)
\]

The particle-scattered flow \( \mathbf{u}^p \) is singular at the particle center and vanishes at infinity. The flow reflected by the interface \( \mathbf{u}^R \) vanishes for \( z' \to \infty \), and the transmitted flow \( \mathbf{u}^T \) vanishes for \( z' \to -\infty \). The asymptotic behavior of the flow fields \( \mathbf{u}^p \) and \( \mathbf{u}^R \) uniquely defines the decomposition (25a).

**b. Scattering, reflection, and transmission operators.** To determine the particle-interface hydrodynamic interactions we introduce operators describing the flow scattered by the particle and the flow reflected or transmitted by the fluid-
fluid interface.

On the particle surface $S$ the two flow components $u^p$ and $u^b$ are related via the no-slip boundary condition

$$u^p + u^b = u^{rb}, \quad r' \in S,$$

where

$$u^{rb} = U + \Omega \times (r' - r) \tag{27}$$

is the rigid-body velocity field corresponding to the particle motion. On the fluid interface $z' = 0$, the flow fields (25a) and (25b) satisfy the velocity-continuity and stress-balance conditions described in Sec. II B.

The boundary conditions and Stokes equations are linear; thus the incoming and reflected flows are linearly related, both for the fluid interface and the particle. We express these linear relations using the operator notation. For a particle, the scattered flow $u^s$ is related to the incoming flow $u^R - u^b$ (measured relative to the particle motion) via the relation

$$u^s = \hat{Z}_p(u^R - u^b), \tag{28}$$

where $\hat{Z}_p$ is the particle scattering operator. Similarly, for the interface we have

$$u^b = \hat{Z}_R u^p, \tag{29}$$

$$u^r = \hat{Z}_T u^p, \tag{30}$$

where $\hat{Z}_R$ and $\hat{Z}_T$ are the interface reflection and transmission operators.

Relation (28) is obtained by solving the Stokes flow problem for a particle in free space; relations (29) and (30) are obtained by solving Stokes equations for a fluid-fluid interface in the absence of the particle. Equations (28)–(30) form a set of linear equations for the unknown fields $u^R$, $u^s$, and $u^r$ for a given rigid-body particle motion [Eq. (27)]. To solve these equations, we project them on complete sets of basis fields, as described in Sec. IV B.

### B. Multipolar and Cartesian expansions

Since the problem involves spherical and planar boundaries, two basic sets of Stokes flows are used: the multipolar set and the Cartesian set. Appropriate transformation relations between these two basis sets are applied to link the multipolar and Cartesian expansions.

The scattering operator $\hat{Z}_p$ describes the interaction of the flow with a spherical particle [cf., Eq. (28)]. The matrix representation $Z_p$ of the operator $\hat{Z}_p$ is thus obtained by expanding the flow fields $u^p$, $u^s$, and $u^b$ into the multipolar basis, consistent with the spherical symmetry of the problem.

The operators $\hat{Z}_R$ and $\hat{Z}_T$ describe flow interaction with the planar interface [cf., Eqs. (29) and (30)]. Their matrix representations $Z_R$ and $Z_T$ are obtained by expanding the flow fields $u^p$, $u^b$, and $u^r$ into the Cartesian basis. After the transformation relations between the two basis sets of Stokes flows have been applied; these expansions reduce the formal operator relations (28)–(30) to a set of algebraic equations that can be solved numerically.

#### a. Multipolar expansion

To determine fluid interaction with the particle, the velocity field $u^p$ (which is singular at the particle center) and the nonsingular velocity fields $u^R$ and $u^{rb}$ are expanded into the corresponding singular $v_{lm}^s$ and nonsingular $v_{lm}^r$ basis fields, introduced in Ref. 41. Accordingly, we have

$$u^p = \sum_{lm} c_p^{lm} v_{lm}^s, \tag{31a}$$

$$u^R = \sum_{lm} c_R^{lm} v_{lm}^r, \tag{31b}$$

$$u^{rb} = \sum_{lm} c_b^{lm} v_{lm}^r, \tag{31c}$$

were $c_p^{lm}$, $c_R^{lm}$, and $c_b^{lm}$ are the expansion coefficients. The flow fields $v_{lm}^r$ are centered at the position of the particle. They depend on the solid angle via the vector spherical harmonics of the order $lm$. The index $\sigma = 0, 1, 2$ corresponds to the pressure, vorticity, and potential solutions. For explicit definitions, see Refs. 8 and 41.

Inserting expansions (31a), (31b), and (31c) into the scattering relation (28) yields a set of algebraic equations for the expansion coefficients. Using a compact matrix notation in the three-dimensional linear space with the components corresponding to the indices $\sigma = 0, 1, 2$, these equations can be written in the form

$$c_{lm}^p = -Z_p(l) \cdot (c_l^R - c_l^{rb}). \tag{32}$$

In the matrix notation, a column vector with components $c_{l\sigma}$ is denoted by $c$, a matrix with elements $A(\sigma|\sigma')$ is denoted by $A$, and the dot represent the matrix product. In the explicit component notation, Eq. (32) is equivalent to

$$c_{lm}^p = -\sum_{\sigma' = 0}^2 Z_p(l; \sigma|\sigma') (c_l^R - c_l^{rb}). \tag{33}$$

In relations (32) and (33), there is no summation over the indices $lm$ because of the spherical symmetry of the particle. Explicit expressions for the elements $Z_p(l; \sigma|\sigma')$ of the particle scattering matrix $Z_p(l)$ are known.8,41

#### b. Cartesian representation

To describe the flow interaction with the interface we use the Cartesian basis of Stokes flows $v_{kor}^s$. According to the expressions listed in the Appendix, the Cartesian basis flows correspond to the lateral Fourier modes (i.e., Fourier modes in the planes parallel to the interface). Similar to the multipolar basis, the indices $\sigma = 0, 1, 2$ describe the pressure, vorticity, and potential solutions of Stokes equations. The fields $v_{kor}^s$ decay exponentially for $z' \to \pm \infty$ [cf., Eq. (A1)].

Taking into account the behavior of basis fields at infinity, we find the following expansions for the particle-scattered flow and the flow components reflected and transmitted by the interface:

$$u^s = \sum_{kor} c_{kor}^s v_{kor}^s, \tag{34a}$$

$$u^r = \sum_{kor} c_{kor}^r v_{kor}^r, \tag{34b}$$

$$u^{rb} = \sum_{kor} c_{kor}^{rb} v_{kor}^r. \tag{34c}$$

The expansion coefficients $c_{kor}^s$, $c_{kor}^r$, and $c_{kor}^{rb}$ can be evaluated by using the transformation matrices $Z_R$ and $Z_T$.

$$c_{kor}^s = \sum_{lm} Z_R(l; \sigma|\sigma') c^R_{lm}, \tag{35a}$$

$$c_{kor}^r = \sum_{lm} Z_T(l; \sigma|\sigma') c^{rb}_{lm}, \tag{35b}$$

$$c_{kor}^{rb} = \sum_{lm} Z_T(l; \sigma|\sigma') c_{lm}^{rb}. \tag{35c}$$

Using Eqs. (28) and (30), the matrix expressions (32)–(35) become

$$c_{lm}^p = -\sum_{\sigma' = 0}^2 Z_p(l; \sigma|\sigma') (c_l^R - c_l^{rb}), \tag{36}$$

$$c_{lm}^r = -\sum_{\sigma' = 0}^2 Z_r(l; \sigma|\sigma') (c_l^R - c_l^{rb}), \tag{37}$$

$$c_{lm}^{rb} = -\sum_{\sigma' = 0}^2 Z_r(l; \sigma|\sigma') (c_l^R - c_l^{rb}). \tag{38}$$

The expressions (36)–(38) provide a compact compact form for the computation of the expansion coefficients $c_{lm}^p$ and $c_{lm}^{rb}$. The expansion coefficient $c_{lm}^r$ can be computed using the inverse of the matrix $Z_r$. The matrix $Z_r$ is singular for $l = 0, 2$, but it can be inverted using the singular value decomposition.

$$Z_r^{-1} = U D V^T,$$

where $U$ and $V$ are orthogonal matrices, and $D$ is a diagonal matrix with the singular values of $Z_r$ on the diagonal. The expansion coefficients $c_{lm}^r$ can be computed using the singular value decomposition of $Z_r$.

$$c_{lm}^r = U D^{-1} V^T (c_l^R - c_l^{rb}).$$

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$$c_{lm}^r = U D^{-1} V^T (c_l^R - c_l^{rb}).$$
\[ \mathbf{u}^p = \int_{\mathbf{k}} \mathbf{d}k \sum_{\sigma} \mathbf{c}^p_{\kappa \sigma} \mathbf{v}_{\kappa \sigma}^+, \quad (34a) \]
\[ \mathbf{u}^r = \int_{\mathbf{k}} \mathbf{d}k \sum_{\sigma} \mathbf{c}^r_{\kappa \sigma} \mathbf{v}_{\kappa \sigma}^-, \quad (34b) \]
\[ \mathbf{u}^T = \int_{\mathbf{k}} \mathbf{d}k \sum_{\sigma} \mathbf{c}^T_{\kappa \sigma} \mathbf{v}_{\kappa \sigma}^+, \quad (34c) \]

where \( \mathbf{c}^p_{\kappa \sigma}, \mathbf{c}^r_{\kappa \sigma}, \) and \( \mathbf{c}^T_{\kappa \sigma} \) are the expansion coefficients. The expansion (34a) converges only for \( z' < z \) because of the singularity of the flow \( \mathbf{u}^p \) at the particle position \( \mathbf{r} \). For \( z' > z \) there exists a corresponding expansion into the basis \( \mathbf{v}_{\kappa \sigma}^- \).

Inserting expressions (34) into the interface reflection and transmission relations (29) and (30) yields the algebraic equations
\[ \mathbf{c}^r_{\kappa} = -\mathbf{Z}_R(\mathbf{k}) \cdot \mathbf{c}^p_{\kappa}, \quad (35) \]
\[ \mathbf{c}^T_{\kappa} = -\mathbf{Z}_T(\mathbf{k}) \cdot \mathbf{c}^p_{\kappa}. \quad (36) \]

In the above equations, different Fourier modes do not couple, owing to the translational symmetry of the interface. Explicit expressions for the reflection matrix \( \mathbf{Z}_R(\mathbf{k}) \) and transmission matrix \( \mathbf{Z}_T(\mathbf{k}) \) for fully compressible and incompressible interfaces are derived in Sec. VI. These matrices are the new key element of our theory.

\textit{c. Transformation relations.} To determine the unknown coefficients \( \mathbf{c}^p_{lm}, \mathbf{c}^r_{lm}, \mathbf{c}^T_{lm}, \) and \( \mathbf{c}^r_{lm} \) in terms of known coefficients \( \mathbf{c}^p_{lm} \) characterizing the rigid-body particle motion, the set of Eqs. (32), (35), and (36) needs to be supplemented by the appropriate transformation relations between the multipolar and Cartesian representations. For our purpose we need the relations
\[ \mathbf{c}^p_{\kappa} = \sum_{lm} \mathbf{T}_{CS}(\mathbf{k},lm) \cdot \mathbf{c}^p_{lm}, \quad (37a) \]
\[ \mathbf{c}^T_{lm} = \int d\mathbf{k} \mathbf{T}_{SC}(lm,\mathbf{k}) \cdot \mathbf{c}^r_{\kappa}. \quad (37b) \]

Explicit expressions for the transformation matrices \( \mathbf{T}_{CS}(\mathbf{k},lm) \) and \( \mathbf{T}_{SC}(lm,\mathbf{k}) \) have been derived in our previous papers.\(^{7,8,43}\) Both \( \mathbf{T}_{CS}(\mathbf{k},lm) \) and \( \mathbf{T}_{SC}(lm,\mathbf{k}) \) depend on \( z \).

\textbf{C. Linear equations for multipolar-expansion coefficients}

Linear equations for the multipolar expansion coefficients \( \mathbf{c}^p_{lm} \) are derived by inverting Eq. (32) and expressing \( \mathbf{c}^r_{lm} \) in terms of \( \mathbf{c}^p_{lm} \) using the wall-reflection relation (35) and transformation relations (37). These algebraic manipulations yield a set of linear equations
\[ \mathbf{Z}_{lm}^p(l) \cdot \mathbf{c}^p_{lm} + \sum_{l'm'} \mathbf{G}'(lm,l'm') \cdot \mathbf{c}^p_{l'm'} = \mathbf{c}^r_{lm}, \quad (38) \]

where
\[ \mathbf{G}'(lm,l'm') = -\int d\mathbf{k} \mathbf{T}_{SC}(lm,\mathbf{k}) \cdot \mathbf{Z}_R(\mathbf{k}) \cdot \mathbf{T}_{CS}(\mathbf{k},l'm'). \quad (39) \]

is the matrix representation of the Green’s operator for Stokes flow in the presence of the interface. \( \mathbf{G}'(lm,l'm') \) depends on the distance from the interface \( z \). The linear Eqs. (38) are diagonal in the spherical-harmonics order \( m, \) i.e.,
\[ \mathbf{G}'(lm,l'm') = \delta_{mm'} \mathbf{G}(lm,l'm), \quad (40) \]

due to the cylindrical symmetry of the problem. Hence, components with different \( m \) decouple and the set of Eqs. (38) can be efficiently solved.

To evaluate the hydrodynamic force acting on the particle, the set of linear Eqs. (38) is solved for a given rigid-body particle motion (described by the expansion coefficients \( \mathbf{c}^p_{lm} \)). Depending on the boundary conditions on the interface, we apply a suitable form of the reflection matrix \( \mathbf{Z}_R(\mathbf{k}) \) (calculated in Sec. VI). The low-order components of the solution \( \mathbf{c}^p_{lm} \) for the flow scattered by the particle are then used to determine the force and torque according to the expressions derived in our earlier publications.\(^{44}\)

\textbf{V. TWO PARTICLES COUPLED THROUGH THE INTERFACE}

The analysis described in Sec. IV will now be generalized to a system with two particles that are placed on the opposite sides of the interface. Particle 1 is in the half space \( z' > 0 \), and particle 2 in the half space \( z' < 0 \). The particles translate and rotate with the linear and angular velocities \( \mathbf{U}_i \) and \( \Omega_i \) \((i = 1, 2)\). We are interested in evaluating the hydrodynamic forces and torques acting on the particles.

\textbf{A. Particle-scattered and wall-reflected flows}

Since the particles are placed on the opposite sides of the interface, their hydrodynamic coupling occurs via the flow transmitted through the interface. To determine this interparticle coupling, we generalize the steps presented in Sec. IV for a single particle.

The two-particle generalization of the flow decomposition (25a) and (25b) is
\[ \mathbf{u} = \mathbf{u}^p + \mathbf{u}^r + \mathbf{u}^T, \quad z' \geq 0; \quad (41a) \]
\[ \mathbf{u} = \mathbf{u}^p + \mathbf{u}^r + \mathbf{u}^T, \quad z' \leq 0, \quad (41b) \]

where the indices \( i = 1 \) and \( 2 \) refer to the particle \( i \). The boundary condition on the surface \( S_i \) of the particle \( i \) thus involves the particle-scattered flow and the flow fields reflected and transmitted by the interface
\[ \mathbf{u}^p + \mathbf{u}^r + \mathbf{u}^T = \mathbf{u}^{rb}_i, \quad \mathbf{r}' \in S_i, \quad (42) \]

where
\[ \mathbf{u}^{rb}_i = \mathbf{U}_i + \Omega_i \times (\mathbf{r}' - \mathbf{r}). \quad (43) \]

In Eq. (42) and in the following expressions, we set \( i = 2 \) for \( i = 1 \), and \( i = 1 \) for \( i = 2 \). Thus \( \mathbf{u}^{rb}_i \) is the flow produced by the
particle present on the opposite side of the interface in relation to the particle \(i\).

The linear boundary condition (42) implies that

\[
\mathbf{u}_i^p = -\mathbf{Z}_p^i(\mathbf{u}_i^p + \mathbf{u}_i^T - \mathbf{u}_i^{r|b}),
\]

(44)

Similarly, for the flows reflected and transmitted by the interface, we obtain

\[
\mathbf{u}_i^r = -\mathbf{Z}_r^i \mathbf{u}_i^p,
\]

(45)

\[
\mathbf{u}_i^T = -\mathbf{Z}_T^i \mathbf{u}_i^p.
\]

(46)

by analogy with relations (28)–(30). The scattering operator \(\mathbf{Z}_p^i\) in Eq. (44) does not depend on the particle position. However, the reflection and transmission operators \(\mathbf{Z}_r^i\) and \(\mathbf{Z}_T^i\) in Eqs. (45) and (46) depend on the half-space \(i\) from which the flow \(\mathbf{u}_i^p\) (incident to the interface) arrives.

B. Multipolar and Cartesian expansions

Similar to the results for the one-particle system, relations (44)–(46) are transformed into a set of linear equations by expanding the component flow fields into the spherical and Cartesian basis fields. By applying multipolar expansions analogous to (31), the scattering relation (44) is transformed into

\[
\mathbf{c}_i^{\text{in}}(\mathbf{r}) = -\mathbf{Z}_p^i(\mathbf{r}) \cdot \left[ \mathbf{c}_i^{\text{R}}(\mathbf{r}) + \mathbf{c}_i^{\text{T}}(\mathbf{r}) - \mathbf{c}_i^{\text{rb}}(\mathbf{r}) \right],
\]

(47)

where \(\mathbf{c}_i^{\text{R}}(\mathbf{r}), \mathbf{c}_i^{\text{in}}(\mathbf{r}), \mathbf{c}_i^{\text{T}}(\mathbf{r}),\) and \(\mathbf{c}_i^{\text{rb}}(\mathbf{r})\) are the expansion coefficients of the flow fields \(\mathbf{u}_i^p, \mathbf{u}_i^r, \mathbf{u}_i^T,\) and \(\mathbf{u}_i^{r|b}\) into the spherical basis flows \(v^+_\text{lms}\) centered at the position of particle \(i\). The corresponding expansions of the reflection and transmission relations (45) and (46) into the Cartesian basis fields yield

\[
\mathbf{c}_i^{\text{R}}(\mathbf{r}) = -\mathbf{Z}_r^i(\mathbf{r}) \cdot \mathbf{c}_i^{\text{R}}(\mathbf{r}),
\]

(48)

\[
\mathbf{c}_i^{\text{T}}(\mathbf{r}) = -\mathbf{Z}_T^i(\mathbf{r}) \cdot \mathbf{c}_i^{\text{T}}(\mathbf{r}).
\]

(49)

We also need the transformation relations

\[
\mathbf{c}_i^{\text{R}}(\mathbf{r}) = \sum_{lm} \mathbf{T}_{\text{RC}}(\mathbf{r};l,\mathbf{k}) \cdot \mathbf{c}_i^{\text{lm}}(\mathbf{r}),
\]

(50a)

\[
\mathbf{c}_i^{\text{R}}(\mathbf{r}) = \int \mathbf{T}_{\text{SC}}(\mathbf{l},\mathbf{k}) \cdot \mathbf{c}_i^{\text{lm}}(\mathbf{r})
\]

(50b)

\[
\mathbf{c}_i^{\text{T}}(\mathbf{r}) = \int \mathbf{T}_{\text{SC}}(\mathbf{l},\mathbf{k}) \cdot \mathbf{c}_i^{\text{lm}}(\mathbf{r})
\]

(50c)

where the matrices \(\mathbf{T}_{\text{RC}}(\mathbf{r};l,\mathbf{k})\) and \(\mathbf{T}_{\text{SC}}(\mathbf{l},\mathbf{k})\), depend on \(\mathbf{r}\), and describe the transformations between the Cartesian representation and the spherical representation centered at particle \(i\). Relations (48)–(50) form a closed set of linear equations for the expansion coefficients.

C. Linear equations for multipolar-expansion coefficients

By analogy with the procedure described in Sec. IV C, all unknown expansion coefficients except for \(\mathbf{c}_i^{\text{rb}}(\mathbf{r})\) can be eliminated from Eqs. (47)–(49) using simple algebraic manipulations. Hence, we obtain a set of linear equations

\[
\left[\mathbf{Z}_p^i(\mathbf{r})\right]^{-1} \cdot \mathbf{c}_i^{\text{in}}(\mathbf{r}) + \sum_{l'm'} \mathbf{G}_1^{ij}(\mathbf{lm},l'm') \cdot \mathbf{c}_{l'm'}^{r|b}(\mathbf{r}) = \mathbf{c}_{l'm'}^{\text{rb}}(\mathbf{r}),
\]

(51a)

\[
\left[\mathbf{Z}_p^i(\mathbf{r})\right]^{-1} \cdot \mathbf{c}_i^{\text{in}}(\mathbf{r}) + \sum_{l'm'} \mathbf{G}_2^{ij}(\mathbf{lm},l'm') \cdot \mathbf{c}_{l'm'}^{r|b}(\mathbf{r}) = \mathbf{c}_{l'm'}^{\text{rb}}(\mathbf{r}).
\]

(51b)

where

\[
\mathbf{G}_1^{ij}(\mathbf{lm},l'm') = -\int d\mathbf{k} \mathbf{T}_{\text{SC}}(\mathbf{k};\mathbf{lm}) \cdot \mathbf{Z}_p^i(\mathbf{k}) \cdot \mathbf{T}_{\text{CS}}(\mathbf{k};l'm'),
\]

(52a)

\[
\mathbf{G}_2^{ij}(\mathbf{lm},l'm') = -\int d\mathbf{k} \mathbf{T}_{\text{SC}}(\mathbf{k};\mathbf{lm}) \cdot \mathbf{Z}_p^i(\mathbf{k}) \cdot \mathbf{T}_{\text{CS}}(\mathbf{k};l'm'),
\]

(52b)

are the Green’s matrices representing hydrodynamic coupling of particle \(i\) to the interface and to the other particle. The forces and torques acting on the particles for a given translational and rotational motion are evaluated from low-order force multipoles obtained from the solution of Eq. (51) by using relations derived previously.\(^{44}\)

For simplicity, our method has been presented here for a two-particle system. However, multiparticle generalization of Eq. (51) can be readily obtained. In addition to the Green’s matrices (52), in such a generalization we also need the Green’s matrix \(\mathbf{G}_{ij}(\mathbf{lm},l'm')\) for two particles \(i\) and \(j\) \((i \neq j)\) at the same side of the interface. As discussed in Refs. 7 and 8, the matrix \(\mathbf{G}_{ij}(\mathbf{lm},l'm')\) includes the direct-interaction contribution \(\mathbf{G}_{ij}^0(\mathbf{lm},l'm')\) and the contribution

\[
\mathbf{G}_{ij}^0(\mathbf{lm},l'm') = -\int d\mathbf{k} \mathbf{T}_{\text{SC}}(\mathbf{k};\mathbf{lm}) \cdot \mathbf{Z}_p^j(\mathbf{k}) \cdot \mathbf{T}_{\text{CS}}(\mathbf{k};l'm'),
\]

(53)

coresponding to the flow scattered from the interface.

VI. REFLECTION AND TRANSMISSION MATRICES

Explicit expressions for the particle-scattering matrix \(\mathbf{Z}_p\) and the transformation matrices \(\mathbf{T}_{\text{SC}}(\mathbf{l},\mathbf{k})\) and \(\mathbf{T}_{\text{CS}}(\mathbf{l},\mathbf{k})\) were given in our previous publications. \(^7,8\) In order to construct the linear Eqs. (38) and (51) we thus only need analytic formulas for the reflection and transmission matrices \(\mathbf{Z}_p\) and \(\mathbf{Z}_r\). In Sec. VI A we derive such expressions for inviscid interfaces that are either fully compressible or incompressible. In Sec. VI B we provide the corresponding formulas for interfaces with nonzero interfacial viscosity.
The results are presented for matrices $Z_R = Z_R^1$ and $Z_T = Z_T^1$, describing reflection and transmission of the incident flow generated in the half-plane $z' > 0$. For the incident flow generated in the half-plane $z' < 0$, the corresponding results for the matrices $Z_R^2$ and $Z_T^2$ are obtained by replacing the viscosity ratio
\[
\lambda = \eta_2 / \eta_1
\]
with $\lambda^{-1} = \eta_1 / \eta_2$.

A. Inviscid interface

1. Fully compressible interface

a. Application of the Lorentz formula. According to the Lorentz formula, Stokes flow in the presence of a planar fluid-fluid interface described by the velocity and stress boundary conditions (5) and (15) can be represented as a combination of the flow in the half-space bounded by a rigid wall and the flow in the presence of a free interface. Application of the Lorentz formula to the reflected and transmitted flows for a given incoming flow $u^R$ yields
\[
\begin{align*}
  u^R &= (1 - \alpha)u^R_{\text{free}} + \alpha u^R_{\text{rw}}, \\
  u^T &= (1 - \alpha)u^T_{\text{free}},
\end{align*}
\]
where
\[
\alpha = \frac{\lambda}{1 + \lambda}
\]
and the subscripts “free” and “rw” refer to the free-interface and rigid-wall solutions. It follows that for a fully compressible interface the matrices $Z_R$ and $Z_T$ have an analogous decomposition
\[
\begin{align*}
  Z_R &= (1 - \alpha)Z^\text{free}_R + \alpha Z^\text{rw}_R, \\
  Z_T &= (1 - \alpha)Z^\text{free}_T.
\end{align*}
\]
Relation (57) allows us to reduce the Stokes-flow problem for a fluid-fluid interface to two simpler problems of Stokes flow in the presence of a free and a rigid interface.

b. Reflection and transmission matrices. The reflection and transmission matrices for a rigid and free interface are obtained using Cartesian-basis definitions (A1)–(A3) and applying appropriate boundary conditions. For a rigid wall, application of the no-slip boundary condition yields
\[
Z^\text{rw}_R = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}.
\]
Relations (35) and (58) indicate that the incoming flow $v^+_k$ produces a reflected flow $v^+_k$ with a unit amplitude.

For a free interface ($\eta_2 = 0$) the normal velocity and the tangential viscous tractions vanish. Application of these conditions yields the reflection matrix
\[
Z^\text{free}_R = \begin{bmatrix} 0 & 0 & 1 \\ 0 & 1 & 0 \\ 1 & 0 & 0 \end{bmatrix}.
\]
The corresponding expression for a transmission matrix is derived by using Eq. (59) and the velocity continuity condition (5)
\[
Z^\text{free}_T = \begin{bmatrix} 1 & 0 & 1 \\ 0 & 2 & 0 \\ 1 & 0 & 1 \end{bmatrix}.
\]
Combining the above relations according to the Lorentz expressions (57), we obtain the following results for the reflection and transmission matrices describing flow interaction with an inviscid compressible interface between fluids with arbitrary viscosities:
\[
\begin{align*}
  Z_R &= \begin{bmatrix} \alpha & 0 & \alpha - 1 \\ 0 & 2\alpha - 1 & 0 \\ \alpha - 1 & 0 & \alpha \end{bmatrix}, \\
  Z_T &= (\alpha - 1) \begin{bmatrix} 1 & 0 & 1 \\ 0 & 2 & 0 \\ 1 & 0 & 1 \end{bmatrix}.
\end{align*}
\]

2. Incompressible interface

a. Surface irrotational and surface solenoidal flows. To obtain the reflection and transmission matrices for an inviscid interface satisfying the incompressibility constraint (18), we use the decomposition of the interfacial flow into the surface irrotational and surface solenoidal components. Specifically, we note that in the plane $z' = 0$ the basis fields (A1) with even values of the index $\sigma$ are surface irrotational
\[
\nabla_s \cdot v^+_k = 0, \quad \nabla_s \times v^+_k = 0, \quad \sigma = 0,2
\]
and those with the odd value of $\sigma$ are surface solenoidal
\[
\nabla_s \cdot v^-_k = 0, \quad \nabla_s \times v^-_k = 0, \quad \sigma = 1.
\]
The corresponding viscous tractions on the interface
\[
\mathbf{f}_k = \mathbf{I}_s \cdot \boldsymbol{\tau}^+_k \cdot \mathbf{e}_z,
\]
where $\boldsymbol{\tau}^+_k$ is the stress tensor associated with the flow $v^+_k$, satisfy the analogous relations
\[
\nabla_s \cdot f^+_k = 0, \quad \nabla_s \times f^+_k = 0, \quad \sigma = 0,2
\]
\[
\nabla_s \cdot f^-_k = 0, \quad \nabla_s \times f^-_k = 0, \quad \sigma = 1.
\]
The above properties of the Cartesian basis flows indicate that the surface irrotational flow components (63) interact with an incompressible interface in the same way as they do with a rigid wall. This can be seen by applying the Helmholtz theorem to the interfacial flow at $z' = 0$. For a combination $u^s$ of the basis fields $v^+_k$ and $v^-_k$, the requirement that
\[
\nabla_s \cdot u^s = 0
\]
in the plane $z' = 0$ implies that...
in this plane because $\mathbf{u}^{\text{irr}}$ is both surface solenoidal and surface irrotational. Hence, we obtain the no-slip boundary condition. The viscous tractions corresponding to the flow field $\mathbf{u}^{\text{irr}}$ are surface irrotational, according to relation (66). Thus they can be balanced by the interfacial tension gradient, consistent with the stress boundary conditions described in Sec. II B.

In contrast, surface tractions $\mathbf{f}_{\text{inter}}^\pm$ with $\sigma=1$ cannot be balanced by the interfacial-tension gradient, according to the relation (67) because they have a nonzero curl. However, flow components $\mathbf{v}_{k1}^\pm$ automatically satisfy the incompressibility condition (18), according to relation (64). Since the flow fields $\mathbf{v}_{k1}^\pm$ do not couple with the interfacial-tension distribution, they interact with an incompressible interface in the same way as they do with a fully compressible interface.

b. Reflection and transmission matrices. The above analysis can be summarized in the following relations for the transmission and reflection matrices for an inviscid incompressible interface

$$
Z_R = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 2\alpha - 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad (70)
$$

$$
Z_T = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 2(\alpha - 1) & 0 \\ 0 & 0 & 0 \end{bmatrix}. \quad (71)
$$

The matrix elements with $\sigma, \sigma'=0,2$ are the same as for a rigid surface [cf., Eq. (58)]; and the matrix elements with $\sigma, \sigma'=1$ are the same as for a compressible interface [cf., Eqs. (61) and (62)]. Moreover, there is no coupling between the subspaces of even and odd indices $\sigma$, which follows from a different parity of the corresponding basis flows.46

B. Viscous interface

For interfaces with nonzero surface viscosity, the Lorentz decomposition (57) does not apply, but the reflection and transmission matrices can be obtained by an explicit calculation.

1. Fully compressible interface

Using expressions (A1) and (A2) for the Cartesian basis fields and applying the velocity boundary condition (5) and stress boundary condition (16) for the fully compressible interface yields

$$
Z_R = \begin{bmatrix} \bar{\alpha} & 0 & \bar{\alpha} - 1 \\ 0 & 2\bar{\alpha} - 1 & 0 \\ \bar{\alpha} - 1 & 0 & \bar{\alpha} \end{bmatrix}, \quad (72)
$$

$$
Z_T = \begin{bmatrix} \bar{\alpha} - 1 & 0 & \bar{\alpha} - 1 \\ 0 & 2(\bar{\alpha} - 1) & 0 \\ \bar{\alpha} - 1 & 0 & \bar{\alpha} - 1 \end{bmatrix}. \quad (73)
$$

where

$$
\bar{\alpha} = \frac{\lambda + \frac{1}{2}(\beta + \kappa)k}{1 + \lambda + \frac{1}{2}(\beta + \kappa)k} \quad (74)
$$

and

$$
\bar{\alpha}' = \frac{\lambda + \beta k}{1 + \lambda + \beta k}. \quad (75)
$$

2. Incompressible interface

For an incompressible interface, the boundary conditions (5), (18), and (21) yield

$$
Z_R = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 2\bar{\alpha}' - 1 & 0 \\ 0 & 0 & 1 \end{bmatrix}, \quad (76)
$$

$$
Z_T = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}. \quad (77)
$$

We note that matrices (72), (73), (76), and (77) depend on the wave vector $k$, and in the long-wavelength limit $k\to0$ they reduce to the corresponding results (61), (62), (70), and (71) for an interface with zero surface viscosity.

The relations for the reflection and transmission matrices derived above, complete our analysis of the hydrodynamic coupling of spherical particles moving in the presence of a fluid-fluid interface. By inserting these relations into (38) and (51) we obtain a closed set of linear equations for the expansion coefficients $c^p_{\text{inter}}$, from which the friction matrix $\zeta$ can be numerically evaluated.

VII. CONCLUSIONS

We have presented a theoretical study of the hydrodynamic coupling between solid spheres and a planar fluid-fluid interface in the creeping-flow regime. Interactions with different types of interfaces have been considered: with compressible and incompressible interfaces, and with interfaces with nonzero surface viscosity. The particles in our analysis can be on the same side or on the opposite sides of the fluid-fluid interface. Our theory combines the HYDROMULTIPOLE expansion technique47 with the Cartesian representation of Stokes flow in the presence of a planar surface.7,8 The numerical algorithm based on our analysis enables highly accurate and efficient calculations for single-particle and multiparticle systems.

In our approach the interactions of the particle with the flow are described using the particle-scattering matrix $Z_p$, and the interactions of the interface with the flow are captured using two matrices, i.e., the reflection matrix $Z_R$ and transmission matrix $Z_T$. The particle-scattering matrices are known for rigid particles,41 particles with stick-slip boundary conditions,48 surfactant-free41 and surfactant-covered56 drops, and for core-shell porous particles.49 The interface reflection matrix was previously evaluated for a rigid wall,7 and the reflection and transmission matrices have been constructed in this paper for a fluid-fluid interface. Our algo-
algorithm can thus be used for a wide range of systems, including spherical drops and polymer-coated porous particles near an interface.

In the accompanying paper we provide numerical results for a single particle in the presence of compressible and incompressible interfaces with no interfacial viscosity. These results have been obtained using the method developed above. The algorithm based on our theoretical analysis can also be applied to determine the mobility of a particle coupled to a fluid-fluid interface with a nonzero interfacial viscosity.

The theoretical approach developed here allows to evaluate hydrodynamic interactions of particles placed on the opposite sides of the interface. We expect that the mutual friction coefficient (or its inverse, the mobility coefficient) in such a system is more sensitive to the interfacial properties than the single-particle friction coefficient. This is because the hydrodynamic interactions between particles separated by the interface are entirely determined by the transmitted flow, and this flow strongly depends on the interfacial properties. Measurements of the mobility coefficient may thus be a sensitive probe of the interfacial properties (such as the surface viscosity).

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APPENDIX: CARTESIAN BASIS OF STOKES FLOWS

The Cartesian basis \( \mathbf{v}_{k_0}(r') \) is given by the following expressions:

\[
\mathbf{v}_{k_0}(r') = k^{-1/2} \mathbf{v}_{k_0}(z') e^{i k \rho' \cdot z'},
\]

where

\[
\rho' = x' \hat{\mathbf{e}}_x + y' \hat{\mathbf{e}}_y,
\]

are the lateral position and wave vectors and

\[
\begin{align*}
\mathbf{V}_{k_0}(z') &= \alpha_0 (i(1 - 2z')) \mathbf{k} + (1 + 2z') \mathbf{e}_z, \\
\mathbf{V}_{k_1}(z') &= 2 \alpha_0 (\mathbf{k} \times \mathbf{e}_z), \\
\mathbf{V}_{k_2}(z') &= \alpha_0 (i \mathbf{k} \mathbf{e}_x), \\
\mathbf{V}_{k_0}(z') &= \alpha_0 (i \mathbf{k} \mathbf{e}_y), \\
\mathbf{V}_{k_1}(z') &= 2 \alpha_0 (\mathbf{k} \mathbf{e}_x), \\
\mathbf{V}_{k_2}(z') &= \alpha_0 (i(1 + 2z') \mathbf{k} - (1 - 2z') \mathbf{e}_z).
\end{align*}
\]

In the above relations, \( k = |\mathbf{k}| \) and \( \mathbf{k} = k / k \) are the length and direction of \( \mathbf{k} \) and \( \alpha_0 = (32 \pi^2)^{-1/2} \) is the normalization coefficient. According to the above equations, the flow fields \( \mathbf{v}_{k_0} \) and \( \mathbf{v}_{k_2} \) are associated with nonzero pressure, \( \mathbf{v}_{k_2} \) and \( \mathbf{v}_{k_2}^* \) are potential flows, and \( \mathbf{v}_{k_1}^* \) are vorticity solutions.

37. In the lubrication regime \( e = \epsilon d / d \), the variation of the viscosity field occurs on the length scale \( \epsilon d / \) and the variation of the interferential tension on the length scale \( \epsilon^2 d / \). Thus, for small particle-interface gaps there is an additional factor \( e^{-1/2} \) on the right-hand side of relations (17) and (22).
41. B. Cichocki, B. U. Felderhof, and R. Schmitz, PCH, PhysicoChem. Hy-
In our previous publications (Refs. 8 and 7), the transformations between the spherical and Cartesian basis fields were decomposed into the transformation matrices and displacement matrices that correspond to a shift of the center of the coordinate system between the particle and wall positions.

Explicit expressions relating the expansion coefficients $c_{lm}^{h}$ and $c_{lm}^{P}$ to the translational and rotational particle velocities and to the force and torque acting on the particle are given in Appendix B of Ref. 7.


