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# Sedimentation of Homogeneous Suspensions in Finite Vessels

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The question of a possible container shape dependence of the sedimentation velocity in a homogeneous suspension is reexamined. To this end we develop a statistical theory of suspensions based on low-Reynolds-number hydrodynamics of spherical particles in a container. It is shown, to first order in the volume fraction, that in an arbitrary vessel the relative sedimentation velocity is shape independent, but that at the same time shape-dependent convection occurs. The theory forms a bridge between earlier calculations for special geometries by Beenakker and Mazur and a phenomenological theory recently proposed by Nozières.

**KEY WORDS**: Sedimentation; long-range hydrodynamic interactions; shape dependence; intrinsic convection.

## 1. INTRODUCTION

Recently some studies have again been devoted to the question of whether the sedimentation of a homogeneous suspension might depend on the shape of its container. Burgers in 1941 stressed the possibility of such a dependence, which he could not rule out, but which did not seem acceptable. This possibility arises from the long range of the so-called hydrodynamic interaction between suspended particles.

These interactions, which result from the fluid flow caused by the motion of a particle, decay very slowly. In fact, in an expansion in inverse powers of the particle separation $R$ they contain, besides shorter ranged contributions, terms of order $R^{-1}$ and $R^{-3}$. The $R^{-1}$ contribution would

This paper is dedicated to N. G. van Kampen.

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lead to a divergence of the sedimentation velocity in an unbounded suspension. This divergence, however, disappears if the interactions with container walls are taken into account, or alternatively, if the relative sedimentation velocity, i.e., the mean particle velocity with respect to the mean volume flow, is considered. But it is the $R^{-3}$ interaction that leads to the question of the shape dependence by giving rise to a conditionally convergent integral in the expression for the relative sedimentation velocity.

In 1972, and again in 1976, Batchelor reconsidered the problem and argued, on the basis of general physical considerations, how this conditionally convergent integral should be evaluated. Since his reasoning is, however, strictly only valid for an unbounded system *without* walls, the question of a possible dependence of sedimentation on the shape of the container could not be considered to be settled definitely.

After Mazur and van Saarloos had formulated a general theory to calculate *many-body* hydrodynamic interactions, Beenakker and Mazur applied this formalism in 1985 to a suspension enclosed in a spherical container. Indeed, even to order linear in the volume fraction $\phi$ of suspended particles, to which only interactions between two suspended particles need be taken into account, one is already faced with a *three-body* problem, with the (spherical) container formally acting as a third particle. It turned out to be possible in this way to calculate to order $\phi$ for a homogeneous suspension and in the center of the spherical container the mean particle velocity as well as the mean volume flow. A similar calculation was also performed for sedimentation toward a plane wall. It was found for both geometries that the *relative* sedimentation has indeed the value found by Batchelor for the unbounded system, but that the mean particle velocity in the laboratory frame had for the spherical container a value different from the one found for sedimentation toward a plane wall. Thus, convection flows depending on shape may occur. These flows represented an unexpected phenomenon for *homogeneous* suspensions, not foreseen in earlier treatments.[3] Of course, this effect is very weak in dilute suspensions, and is moreover rapidly masked by the much larger buoyancy-driven convection occurring if inhomogeneities have been induced during sedimentation, for instance, by inclined walls. This, however, does not imply that the effect has no relevance: after all, its strength has the same order of magnitude as the intensively studied (first-order) density correction to the sedimentation coefficient.

[3] In fact, in his 1976 paper Batchelor explicitly states that for a collection of particles, each of which is acted on by the same steady force, "the average particle velocity [relative to the mean volume flow]... is of course [equal to] the average particle velocity relative to the walls of a vessel containing a statistically homogeneous suspension." In other words, he states that the mean volume flow relative to the walls of the vessel is zero.

Nozières recently showed that the above-mentioned rather surprising phenomenon could be understood on a "macroscopic" level (i.e., a level on which the suspended particles have lost their identity, so to say) as a consequence of the *local* coupling between two flow fields, the relative sedimentation velocity and the mean volume velocity. He formulated, with profound physical insight, a theory of sedimentation in terms of two coupled differential equations for these fields containing a number of phenomenological coefficients and supplemented by an "effective" boundary condition. The solutions of Nozières' equations contain the shape-independent relative sedimentation velocity as well as the shape-dependent intrinsic convection phenomenon found for a spherical container geometry by Beenakker and Mazur.

In this paper we present a "microscopic" derivation of a "macroscopic" sedimentation theory for a suspension of spherical particles contained in a vessel of arbitrary shape. The theory is given up to linear order in the volume fraction $\phi$. In Section 2 we give, within the framework of a formalism of induced forces, the formal solution of the linearized Stokes equation for a system of spheres moving in a fluid within a container. From this solution we derive in Section 3, by averaging over particle configurations, to linear order in $\phi$, general expressions for the sedimentation velocity and the volume flow.

In Section 4 we discuss, for a better understanding of the problem, the unbounded suspension. The expression for the sedimentation velocity then contains the conditionally convergent integral mentioned above. We show that a relation exists which allows us to express this conditionally convergent integral in terms of the Laplacian of the volume velocity. Batchelor's argument to resolve the problem of a possible shape dependence in sedimentation amounts to stating that the latter quantity must be zero on physical grounds, i.e., for reasons of symmetry. Then, due to the relation found, a particular value must be attributed to the conditionally convergent integral. For a finite system, however, this integral has a value which does depend on the shape of the container. Since nevertheless the relative sedimentation velocity is expected to be shape independent, for a finite system there must be other, compensating, shape-dependent integrals in the expression for the relative sedimentation velocity. On the other hand, in view of the previous results pointing to the existence of the phenomenon of "essential" or "intrinsic" convection, the mean volume velocity must obey an equation which has shape-dependent solutions, however large the vessel may be.

These points are explicitly shown in Section 5. We also demonstrate in this section that on the "macroscopic" scale the bulk part of the solution for the volume velocity $\mathbf{V}$ corresponds to the solution which would be found from the differential equation for $\mathbf{V}$ valid in the bulk part of the system, with an effective boundary condition at the container surface. As Nozières already surmised, the value of the phenomenological coefficient in this boundary condition inferred from the Beenakker–Mazur result constitutes only a lowest order approximation.

The microscopic theory for arbitrary container shape presented in Section 5, which explicitly takes into account the hydrodynamic interactions with container walls, may be considered to provide a justification for Nozières' phenomenological approach to sedimentation.

In the rather technical Section 6 we discuss in detail that surface corrections to Batchelor's value for the relative sedimentation velocity and to Nozières' equations indeed vanish as the surface recedes to infinity.

Finally, we make some concluding remarks.

## 2. HYDRODYNAMIC EQUATIONS

We consider a system of $N$ spherical particles of common radius $a$ and with centers at $\mathbf{R}_1,\ldots,\mathbf{R}_N$ suspended in an incompressible fluid of viscosity $\eta$ and enclosed in a container of volume $\mathscr{V}$. The pressure tensor $\mathsf{P}(\mathbf{r})$ in the fluid is given in terms of the velocity field $\mathbf{v}(\mathbf{r})$ and the pressure field $p(\mathbf{r})$ by

$$P_{\alpha\beta}(\mathbf{r}) = \delta_{\alpha\beta}\, p(\mathbf{r}) - \eta\left(\frac{\partial v_\alpha}{\partial r_\beta} + \frac{\partial v_\beta}{\partial r_\alpha}\right) \tag{2.1}$$

and obeys the quasistatic Stokes equation

$$-\nabla\cdot\mathsf{P}(\mathbf{r}) + \rho_f\mathbf{g} = 0 \qquad \text{for} \quad |\mathbf{r}-\mathbf{R}_i| > a, \quad i=1,\ldots,N \tag{2.2}$$

Here $\rho_f$ denotes the fluid mass density and $\mathbf{g}$ the earth's gravitational field. We employ stick boundary conditions. On the surface $W$ of the container we thus have

$$\mathbf{v}(\mathbf{r}) = 0 \qquad (\mathbf{r}\in W) \tag{2.3}$$

whereas on the surfaces of the particles

$$\mathbf{v}(\mathbf{r}) = \mathbf{u}_i + \boldsymbol{\omega}_i \wedge (\mathbf{r}-\mathbf{R}_i) \qquad (|\mathbf{r}-\mathbf{R}_i| = a) \tag{2.4}$$

holds, with $\mathbf{u}_i$ and $\boldsymbol{\omega}_i$ the translational and angular velocities of sphere $i$, respectively.

Due to stationarity the total force acting on each sphere is zero. In other words, the hydrodynamic force cancels the gravitational force on each sphere,

$$-\int \hat{\mathbf{n}}\cdot\mathsf{P}(\mathbf{R}_i + a\hat{\mathbf{n}})\, a^2\, d\hat{\mathbf{n}} + \frac{4\pi}{3}a^3\rho_s\mathbf{g} = 0 \tag{2.5}$$

where $\rho_s$ is the mass density of the spheres. $\hat{\mathbf{n}}$ denotes a unit vector and $\int d\hat{\mathbf{n}} \cdots$ integration over the surface of the unit sphere. We note that the gravitational field exerts no torque on the particles. Therefore, due to stationarity, the torques exerted by the fluid on the particles also vanish.

As in previous work, a reformulation of the flow problem in terms of induced forces is convenient. The fluid equation of motion (2.1), (2.2) can then be extended to all of space and written in the form

$$-\nabla \cdot \mathsf{P}(\mathbf{r}) = \eta\, \Delta \mathbf{v}(\mathbf{r}) - \nabla p(\mathbf{r}) = -\sum_{i=0}^{N} \mathbf{F}_i(\mathbf{r} \mid \mathbf{R}_1,..., \mathbf{R}_N, W) - \chi_W(\mathbf{r})\, \rho_f \mathbf{g} \tag{2.6}$$

*for all* $\mathbf{r}$; the function $\chi_W(\mathbf{r})$ is unity for $\mathbf{r}$ inside and zero for $\mathbf{r}$ outside the container. Equation (2.6) is equivalent to the original boundary value problem constituted by Eqs. (2.1)–(2.4) if one imposes the following requirements on the induced forces $\mathbf{F}_i$ and the extensions of the pressure and velocity fields (cf., e.g., ref. 1):

$$\mathbf{F}_i(\mathbf{r} \mid \mathbf{R}_1,..., \mathbf{R}_N, W) = 0 \qquad \text{for} \quad |\mathbf{r} - \mathbf{R}_i| > a \tag{2.7a}$$

$$p(\mathbf{r}) = \rho_f \mathbf{g} \cdot \mathbf{r} \qquad \text{for} \quad |\mathbf{r} - \mathbf{R}_i| < a, \quad i = 1,..., N \tag{2.7b}$$

$$\mathbf{v}(\mathbf{r}) = \mathbf{u}_i + \boldsymbol{\omega}_i \wedge (\mathbf{r} - \mathbf{R}_i) \quad \text{for} \quad |\mathbf{r} - \mathbf{R}_i| \leqslant a \tag{2.7c}$$

$$\mathbf{F}_0(\mathbf{r} \mid \mathbf{R}_1,..., \mathbf{R}_N, W) = 0 \qquad \text{for } \mathbf{r} \text{ inside the container} \tag{2.8a}$$

$$p(\mathbf{r}) = 0 \qquad \text{for } \mathbf{r} \text{ outside the container} \tag{2.8b}$$

$$\mathbf{v}(\mathbf{r}) = 0 \qquad \text{for } \mathbf{r} \text{ outside the container and on its wall} \tag{2.8c}$$

The induced forces depend on the positions $\mathbf{R}_1,..., \mathbf{R}_N$ of the centers of all spheres as well as on the set $W$ of all points of the container wall; for the sake of brevity we often omit the arguments $\mathbf{R}_1,..., W$.

As a consequence of these extensions, the induced force densities on the spheres are of the form

$$\mathbf{F}_i(\mathbf{r}) = \mathbf{f}_i\left(\frac{\mathbf{r} - \mathbf{R}_i}{a}\right) \delta(|\mathbf{r} - \mathbf{R}_i| - a) \qquad (i = 1,..., N) \tag{2.9}$$

Similarly, the induced force density $\mathbf{F}_0$ is concentrated as a surface force density $\mathbf{f}_0$ on the container wall.

Next we introduce a reduced pressure $p'(\mathbf{r})$ by

$$p'(\mathbf{r}) = \begin{cases} p(\mathbf{r}) - \rho_f \mathbf{g} \cdot \mathbf{r} & \text{inside the container} \\ 0 & \text{outside the container} \end{cases} \tag{2.10}$$

Equation (2.6) then takes the form

$$\eta\,\Delta\mathbf{v}(\mathbf{r})-\nabla p'(\mathbf{r})=-\sum_{i=0}^{N}\mathbf{F}_i(\mathbf{r}) \tag{2.11}$$

and must be solved with the subsidiary incompressibility condition

$$\nabla\cdot\mathbf{v}(\mathbf{r})=0 \tag{2.12}$$

Using the Oseen tensor $\mathsf{T}(\mathbf{r})\equiv(1/r)(\mathsf{1}+\hat{\mathbf{r}}^2)$, with $\mathsf{1}$ the unit tensor of rank 2 and $\hat{\mathbf{r}}^2$ the dyadic product $\hat{\mathbf{r}}\hat{\mathbf{r}}$, one finds[(2)]

$$\begin{aligned}\mathbf{v}(\mathbf{r})=&\sum_{i=1}^{N}\frac{1}{8\pi\eta}\int\mathsf{T}(\mathbf{r}-\mathbf{R}_i-a\hat{\mathbf{n}})\cdot\mathbf{f}_i(\hat{\mathbf{n}})\,a^2\,d\hat{\mathbf{n}}\\&+\frac{1}{8\pi\eta}\int_W\mathsf{T}(\mathbf{r}-\mathbf{r}')\cdot\mathbf{f}_0(\mathbf{r}')\,ds'\end{aligned} \tag{2.13}$$

where $ds'$ is the surface element on the container wall. With the aid of the integral kernel

$$\Pi(\mathbf{r})\equiv-\frac{3}{4\pi}\frac{1}{r^2}\hat{\mathbf{r}}^3 \tag{2.14}$$

one can express the modified pressure tensor

$$\mathsf{P}'(\mathbf{r})\equiv\mathsf{P}(\mathbf{r})+[p'(\mathbf{r})-p(\mathbf{r})]\mathsf{1} \tag{2.15}$$

in an analogous way[4]

$$\begin{aligned}\mathsf{P}'(\mathbf{r})=&\sum_{i=1}^{N}\int\Pi(\mathbf{r}-\mathbf{R}_i-a\hat{\mathbf{n}})\cdot\mathbf{f}_i(\hat{\mathbf{n}})\,a^2\,d\hat{\mathbf{n}}\\&+\int_W\Pi(\mathbf{r}-\mathbf{r}')\cdot\mathbf{f}_0(\mathbf{r}')\,ds'\end{aligned} \tag{2.16}$$

We conclude this section by establishing a relation between the induced force $\mathbf{F}_i$ and the gravitational force on sphere $i$. To this end, we apply Gauss' theorem to Eq. (2.5) to obtain

$$\begin{aligned}0&=-\int\hat{\mathbf{n}}\cdot\mathsf{P}(\mathbf{R}_i+a\hat{\mathbf{n}})a^2\,d\hat{\mathbf{n}}+\frac{4\pi}{3}a^3\rho_s\,\mathbf{g}\\&=-\lim_{\varepsilon\downarrow 0}\int_{r<a+\varepsilon}\nabla\cdot\mathsf{P}(\mathbf{R}_i+\mathbf{r})\,d\mathbf{r}+\frac{4\pi}{3}a^3\rho_s\,\mathbf{g}\\&=-\lim_{\varepsilon\downarrow 0}\int_{r<a+\varepsilon}\{\mathbf{F}_i(\mathbf{r})+\rho_f\,\mathbf{g}\}\,d\mathbf{r}+\frac{4\pi}{3}a^3\rho_s\,\mathbf{g}\end{aligned} \tag{2.17}$$

[4] This may be verified by solving the Poisson equation $\nabla\cdot(\eta\,\Delta\mathbf{v}-\nabla\mathbf{p}')=-\Delta\mathbf{p}'=-\sum_i\nabla\cdot\mathbf{F}_i$ for $p$ and using Eq. (2.13) to calculate $(\nabla\mathbf{v})^s$, the symmetric velocity gradient.

Thus, the total induced force on sphere $i$ is equal to the buoyancy-corrected gravitational force on it. Since we consider a stationary situation, this force must be compensated by the friction force **K** which the fluid exerts on the sphere,

$$\mathbf{K} = -\int \mathbf{F}_i(\mathbf{r})\, d\mathbf{r} = -\frac{4\pi}{3} a^3(\rho_s - \rho_f)\mathbf{g} \tag{2.18}$$

## 4. GENERAL FORMULAS FOR SEDIMENTATION VELOCITY AND VOLUME FLOW

We now consider the motion of a sphere $i$ at position $\mathbf{R}_i$. In order to express its translational velocity $\mathbf{u}_i$ in terms of the induced forces, we integrate the boundary condition (2.4) over the surface of sphere $i$ and substitute relation (2.13),

$$\begin{aligned}\mathbf{u}_i &= \frac{1}{4\pi}\int d\hat{\mathbf{n}}\, \{\mathbf{u}_i + \boldsymbol{\omega}_i \wedge (\hat{\mathbf{n}}a + \mathbf{R}_i)\} \\ &= \frac{1}{4\pi}\int d\hat{\mathbf{n}}\, \mathbf{v}(\mathbf{R}_i + a\hat{\mathbf{n}}) \\ &= \frac{1}{4\pi}\int d\hat{\mathbf{n}}\, \frac{1}{8\pi\eta}\left\{\sum_{j=1}^{N}\int d\hat{\mathbf{n}}'\, a^2 \mathsf{T}(\mathbf{R}_i + a\hat{\mathbf{n}} - \mathbf{R}_j - a\hat{\mathbf{n}}')\cdot \mathbf{f}_j(\hat{\mathbf{n}}')\right. \\ &\quad \left. + \int_W ds'\, \mathsf{T}(\mathbf{R}_i + a\hat{\mathbf{n}} - \mathbf{r}')\cdot \mathbf{f}_0(\mathbf{r}')\right\}\end{aligned} \tag{3.1}$$

To proceed, we need the integral relation

$$\frac{3a}{4}\frac{1}{4\pi}\int d\hat{\mathbf{n}}\, \mathsf{T}(\mathbf{R} + a\hat{\mathbf{n}}) = \begin{cases} \frac{3}{4}a\mathsf{T}(\mathbf{R}) + \frac{1}{4}a^3 \mathsf{D}(\mathbf{R}) \equiv \mathsf{A}(\mathbf{R}) & \text{for } R \geqslant a \\ \mathsf{1} & \text{for } R < a \end{cases} \tag{3.2}$$

with

$$\mathsf{D}(\mathbf{R}) \equiv \frac{1}{R^3}(\mathsf{1} - 3\hat{\mathbf{R}}^2) \qquad (R > 0) \tag{3.3}$$

For later use we note here that

$$\frac{1}{4\pi}\int d\hat{\mathbf{n}}\, \mathsf{D}(\mathbf{R} + a\hat{\mathbf{n}}) = \begin{cases} \mathsf{D}(\mathbf{R}) & \text{for } R > a \\ 0 & \text{for } R < a \end{cases} \tag{3.4}$$

Using formula (3.2), we find from (3.1)

$$6\pi\eta a \mathbf{u}_i = -\mathbf{K} + \sum_{\substack{j=1 \\ j\neq i}}^{N} \int d\hat{\mathbf{n}}\, a^2 \mathsf{A}(\mathbf{R}_i - \mathbf{R}_j - a\hat{\mathbf{n}}) \cdot \mathbf{f}_j(\hat{\mathbf{n}}) + \int_W ds'\, \mathsf{A}(\mathbf{R}_i - \mathbf{r}') \cdot \mathbf{f}_0(\mathbf{r}') \tag{3.5}$$

For our purpose it is necessary to split the force density $\mathbf{f}_j(\hat{\mathbf{n}})$ into a constant part and a part $\mathbf{h}_j(\hat{\mathbf{n}})$ in such a way that the integral over $\mathbf{h}_j(\hat{\mathbf{n}})$ vanishes. $\mathbf{h}_j(\hat{\mathbf{n}})$ is thus defined by

$$\mathbf{f}_j(\hat{\mathbf{n}}) = \mathbf{h}_j(\hat{\mathbf{n}}) + \frac{1}{4\pi} \int d\hat{\mathbf{n}}\, \mathbf{f}_j(\hat{\mathbf{n}}) = \mathbf{h}_j(\hat{\mathbf{n}}) - \frac{1}{4\pi a^2} \mathbf{K} \tag{3.6}$$

In the last member of this equation use was made of the relation (2.18). After substitution of formula (3.6) into Eq. (3.5) the integrals containing the constant parts $-(1/4\pi a^2)\mathbf{K}$ of the force densities $\mathbf{f}_j$ can be evaluated:

$$6\pi\eta a \mathbf{u}_i = -\mathbf{K} + \sum_{\substack{j=1 \\ j\neq i}}^{N} \left\{ -\left[\frac{3}{4} a \mathsf{T}(\mathbf{R}_i - \mathbf{R}_j) + \frac{a^3}{2} \mathsf{D}(\mathbf{R}_i - \mathbf{R}_j)\right] \cdot \mathbf{K} + \int d\hat{\mathbf{n}}\, a^2 \mathsf{A}(\mathbf{R}_i - \mathbf{R}_j - a\hat{\mathbf{n}}) \cdot \mathbf{h}_j \hat{\mathbf{n}}) \right\} + \int_W ds'\, \mathsf{A}(\mathbf{R}_i - \mathbf{r}') \cdot \mathbf{f}_0(\mathbf{r}') \tag{3.7}$$

where use has been made of formulas (3.2)–(3.4). In particular, when only two particles are present, Eq. (3.7) takes the form

$$\begin{aligned} 6\pi\eta a \mathbf{u}_1 = & -\mathbf{K} + \int_W ds'\, \mathsf{A}(\mathbf{R}_1 - \mathbf{r}') \cdot \mathbf{f}_0(\mathbf{r}' \mid \mathbf{R}_1, W) \\ & - \left[\frac{3}{4} a \mathsf{T}(\mathbf{R}_1 - \mathbf{R}_2) + \frac{a^3}{2} \mathsf{D}(\mathbf{R}_1 - \mathbf{R}_2)\right] \cdot \mathbf{K} \\ & + \int d\hat{\mathbf{n}}\, a^2 \mathsf{A}(\mathbf{R}_1 - \mathbf{R}_2 - a\hat{\mathbf{n}}) \cdot \mathbf{h}_2(\hat{\mathbf{n}} \mid \mathbf{R}_1, \mathbf{R}_2, W) \\ & + \int_W ds'\, \mathbf{A}(\mathbf{R}_1 - \mathbf{r}') \cdot [\mathbf{f}_0(\mathbf{r}' \mid \mathbf{R}_1, \mathbf{R}_2, W) - \mathbf{f}_0(\mathbf{r}' \mid \mathbf{R}_1, W)] \end{aligned} \tag{3.8}$$

The first two terms on the rhs are the single-particle contribution to $\mathbf{u}_1$; they remain unchanged if particle 2 is removed. The other terms represent the modification of $\mathbf{u}_1$ due to the presence of the second particle.

We now evaluate for a homogeneous suspension the average velocity $\mathbf{U}(\mathbf{R}_1)$ of a particle with center at $\mathbf{R}_1$. We restrict the discussion to the case of a dilute suspension, in which case it suffices to take into account only terms linear in the volume fraction $\phi = \frac{4}{3}\pi a^3 N/\mathscr{V}$. To first order in $\phi$ the nonadditivity of hydrodynamic interactions plays no role. One may then write the many-particle interaction contribution to $\mathbf{U}(\mathbf{R}_1)$ as a superposition of two-particle terms. The conditional probability density $P(\mathbf{R}_2 \mid \mathbf{R}_1)$ for finding a particle at $\mathbf{R}_2$, given that there is a particle at $\mathbf{R}_1$, is given to this order by

$$P(\mathbf{R}_2 \mid \mathbf{R}_1) = \begin{cases} 1/\mathscr{V} & \text{for} \quad |\mathbf{R}_1 - \mathbf{R}_2| > 2a, \quad d(\mathbf{R}_2) > a \\ 0 & \text{elsewhere} \end{cases} \tag{3.9}$$

where $d(\mathbf{R})$ denotes the distance of the point $\mathbf{R}$ inside the container to the container wall.

One thus finds from (3.8) for the average particle velocity

$$\begin{aligned} 6\pi\eta a \mathbf{U}(\mathbf{R}_1) = & -\mathbf{K} + \int_W ds' \, \mathsf{A}(\mathbf{R}_1 - \mathbf{r}') \cdot \mathbf{f}_0(\mathbf{r}' \mid \mathbf{R}_1, W) \\ & - \frac{N}{\mathscr{V}} \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a \\ d(\mathbf{R}_2) > a}} d\mathbf{R}_2 \frac{3a}{4} \mathsf{T}(\mathbf{R}_1 - \mathbf{R}_2) \cdot \mathbf{K} \\ & - \frac{N}{\mathscr{V}} \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a \\ d(\mathbf{R}_2) > a}} d\mathbf{R}_2 \frac{a^3}{2} \mathsf{D}(\mathbf{R}_1 - \mathbf{R}_2) \cdot \mathbf{K} \\ & + \frac{N}{\mathscr{V}} \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a \\ d(\mathbf{R}_2) > a}} d\mathbf{R}_2 \left\{ \int d\hat{\mathbf{n}} \, a^2 \mathsf{A}(\mathbf{R}_1 - \mathbf{R}_2 - a\hat{\mathbf{n}}) \cdot \mathbf{h}_2(\hat{\mathbf{n}} \mid \mathbf{R}_1, \mathbf{R}_2, W) \right. \\ & \left. + \int_W ds' \mathsf{A}(\mathbf{R}_1 - \mathbf{r}') \cdot [\mathbf{f}_0(\mathbf{r}' \mid \mathbf{R}_1, \mathbf{R}_2, W) - \mathbf{f}_0(\mathbf{r}' \mid \mathbf{R}_1, W)] \right\} \end{aligned} \tag{3.10}$$

The second integral on the right-hand side diverges in the limit of an infinitely large container. This divergent contribution to $\mathbf{U}$ is, however, compensated by a backflow generated by the induced forces on the container wall,[3,4] a fact which clearly demonstrates that the influence of walls persists even in the limit of infinite container dimensions. For the infinite system *without* wall the difficulty arising from the divergent integral, the so-called Smoluchowski paradox, may be avoided[5] by considering sedimentation with respect to the mean volume flow $\mathbf{V}(\mathbf{r})$. This relative sedimentation velocity $\mathbf{w}(\mathbf{r}) \equiv \mathbf{U}(\mathbf{r}) - \mathbf{V}(\mathbf{r})$ is the proper quantity to describe sedimentation also when container walls are present, because, *in contrast* to $\mathbf{U}(\mathbf{r})$, it will turn out to be *independent of the container shape.*

The mean volume flow $\mathbf{V}(\mathbf{r})$ can be found by taking the average of the velocity field $\mathbf{v}(\mathbf{r})$ occurring in Eq. (2.6), with the extension defined in Eq. (2.7c), over all configurations of the spheres. Indeed, the velocity field $\mathbf{v}(\mathbf{r})$ gives the actual physical velocity of the point $\mathbf{r}$, regardless of whether this point is inside a particle or inside the fluid. To lowest order in $\phi$ the mean of $\mathbf{v}(\mathbf{r})$ is obtained by averaging Eq. (2.13) with induced forces corresponding to the case that there is only one particle. In this way one finds, using Eq. (3.2), for $d(\mathbf{r}) > 2a$,

$$\begin{aligned}
6\pi\eta a\mathbf{V}(\mathbf{r}) &= \frac{N}{\mathscr{V}}\int_{d(\mathbf{R}_2)>a} d\mathbf{R}_2 \left\{ \int d\hat{\mathbf{n}}\, a^2 \frac{3a}{4} \mathsf{T}(\mathbf{r}-\mathbf{R}_2-a\hat{\mathbf{n}}) \right. \\
&\qquad \cdot \left[ -\frac{\mathbf{K}}{4\pi a^2} + \mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_2, W) \right] \\
&\qquad \left. + \int_W ds' \frac{3a}{4} \mathsf{T}(\mathbf{r}-\mathbf{r}')\cdot \mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_2, W) \right\} \\
&= -\phi\mathbf{K} - \frac{N}{\mathscr{V}} \int_{\substack{|\mathbf{R}_2-\mathbf{r}|>a \\ d(\mathbf{R}_2)>a}} \left[ \frac{3}{4} a\mathsf{T}(\mathbf{r}-\mathbf{R}_2) + \frac{a^3}{4}\mathsf{D}(\mathbf{r}-\mathbf{R}_2) \right]\cdot\mathbf{K} \\
&\quad + \frac{N}{V}\int_{d(\mathbf{R}_2)>a} d\mathbf{R}_2 \left\{ \int d\hat{\mathbf{n}}\, a^2 \frac{3a}{4} \mathsf{T}(\mathbf{r}-\mathbf{R}_2-a\hat{\mathbf{n}})\cdot \boldsymbol{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_2, W) \right. \\
&\qquad \left. + \int_W ds' \frac{3a}{4} \mathsf{T}(\mathbf{r}-\mathbf{r}')\cdot \mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_2, W) \right\}
\end{aligned} \tag{3.11}$$

Subtraction of the mean volume flow at $\mathbf{r} = \mathbf{R}_1$ from the sedimentation velocity $\mathbf{U}(\mathbf{R}_1)$ [cf. Eq. (3.10)] yields for the relative sedimentation velocity for $d(\mathbf{R}_1) > 2a$ the result

$$\begin{aligned}
&6\pi\eta a\mathbf{w}(\mathbf{R}_1) \\
&\quad = -(1-\phi)\mathbf{K} + \int_W ds'\, \mathsf{A}(\mathbf{R}_1-\mathbf{r}')\cdot\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_1, W) \\
&\qquad + \frac{N}{\mathscr{V}} \int_{\substack{a<|\mathbf{R}_1-\mathbf{R}_2|<2a \\ d(\mathbf{R}_2)>a}} \int d\mathbf{R}_2\, \mathsf{A}(\mathbf{R}_1-\mathbf{R}_2)\cdot\mathbf{K} \\
&\qquad - \frac{N}{\mathscr{V}} \int_{\substack{|\mathbf{R}_1-\mathbf{R}_2|>2a \\ d(\mathbf{R}_2)>a}} d\mathbf{R}_2 \frac{a^3}{4} \mathsf{D}(\mathbf{R}_1-\mathbf{R}_2)\cdot\mathbf{K}
\end{aligned}$$

$$+\frac{N}{\mathscr{V}}\int_{\substack{|\mathbf{R}_1-\mathbf{R}_2|>2a\\ d(\mathbf{R}_2)>a}}\left\{\int d\hat{\mathbf{n}}\, a^2 \mathsf{A}(\mathbf{R}_1-\mathbf{R}_2-a\hat{\mathbf{n}})\right.$$

$$\cdot[\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_1,\mathbf{R}_2,W)-\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_2,W)]+\int_W ds'\,\mathsf{A}(\mathbf{R}_1-\mathbf{r}')$$

$$\left.\cdot[\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_1,\mathbf{R}_2,W)-\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_1,W)-\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_2,W)]\right\}$$

$$+\frac{N}{\mathscr{V}}\int_{\substack{|\mathbf{R}_1-\mathbf{R}_2|>2a\\ d(\mathbf{R}_2)>a}}\left\{\int d\hat{\mathbf{n}}\,a^2\frac{a^3}{4}\mathsf{D}(\mathbf{R}_1-\mathbf{R}_2-a\hat{\mathbf{n}})\cdot\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_2,W)\right.$$

$$\left.+\int_W ds'\,\frac{a^3}{4}\mathsf{D}(\mathbf{R}_1-\mathbf{r}')\cdot\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_2,W)\right\}$$

$$-\frac{N}{\mathscr{V}}\int_{\substack{|\mathbf{R}_1-\mathbf{R}_2|<2a\\ d(\mathbf{R}_2)>a}}\left\{\int d\hat{\mathbf{n}}\,a^2\frac{3a}{4}\mathsf{T}(\mathbf{R}_1-\mathbf{R}_2-a\hat{\mathbf{n}})\cdot\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_2,W)\right.$$

$$\left.+\int_W ds'\,\frac{3a}{4}\mathsf{T}(\mathbf{R}_1-\mathbf{r}')\cdot f_0(\mathbf{r}'\,|\,\mathbf{R}_2,W)\right\} \tag{3.12}$$

The second integral on the right-hand side can be performed explicitly, using formulas (3.2)–(3.4), and one finds, for $d(\mathbf{R}_1)>3a$,[5]

$$6\pi\eta a\mathbf{w}(\mathbf{R}_1)$$

$$=-\left(1-\frac{11}{2}\phi\right)\mathbf{K}+\int_W ds'\,\mathsf{A}(\mathbf{R}_1-\mathbf{r}')\cdot\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_1,W)$$

$$-\frac{N}{\mathscr{V}}\int_{\substack{|\mathbf{R}_1-\mathbf{R}_2|>2a\\ d(\mathbf{R}_2)>a}} d\mathbf{R}_2\,\frac{a^3}{4}\mathsf{D}(\mathbf{R}_1-\mathbf{R}_2)\cdot\mathbf{K}$$

$$+\frac{N}{\mathscr{V}}\int_{\substack{|\mathbf{R}_1-\mathbf{R}_2|>2a\\ d(\mathbf{R}_2)>a}} d\mathbf{R}_2\int d\hat{\mathbf{n}}a^2\mathsf{A}(\mathbf{R}_1-\mathbf{R}_2-a\hat{\mathbf{n}})\cdot\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_1,\mathbf{R}_2)$$

$$+\frac{N}{\mathscr{V}}\int_{\substack{|\mathbf{R}_1-\mathbf{R}_2|>2a\\ d(\mathbf{R}_2)>a}} d\mathbf{R}_2\left\{\int d\hat{\mathbf{n}}\,a^2\mathsf{A}(\mathbf{R}_1-\mathbf{R}_2-a\hat{\mathbf{n}})\right.$$

$$\cdot[\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_1,\mathbf{R}_2,W)-\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_2,W)-\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_1,\mathbf{R}_2)]$$

$$\left.+\int_W ds'\mathsf{A}(\mathbf{R}_1-\mathbf{r}')\cdot[\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_1,\mathbf{R}_2,W)-\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_1,W)-\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_2,W)]\right\}$$

[5] If the distance from $\mathbf{R}_1$ to the wall is smaller than $3a$, the term $(11/2)\,\phi\mathbf{K}$ on the rhs of Eq. (3.13) is modified and depends explicitly on this distance. We are, however, not interested in the *sedimentation velocity* that close to the wall.

$$+\frac{N}{\mathscr{V}}\int_{\substack{|\mathbf{R}_1-\mathbf{R}_2|>2a\\ d(\mathbf{R}_2)>a}}\left\{\int d\hat{\mathbf{n}}\, a^2\frac{a^3}{4}\,\mathsf{D}(\mathbf{R}_1-\mathbf{R}_2-a\hat{\mathbf{n}})\cdot\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_2, W)\right.$$

$$\left.+\int_W ds'\frac{a^3}{4}\,\mathsf{D}(\mathbf{R}_1-\mathbf{r}')\cdot\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_2, W)\right\}$$

$$-\frac{N}{\mathscr{V}}\int_{\substack{|\mathbf{R}_1-\mathbf{R}_2|<2a\\ d(\mathbf{R}_2)>a}} d\mathbf{R}_2\left\{\int d\hat{\mathbf{n}}\, a^2\frac{3a}{4}\,\tau(\mathbf{R}_1-\mathbf{R}_2-a\hat{\mathbf{n}})\cdot\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_2, W)\right.$$

$$\left.+\int_W ds'\frac{3a}{4}\,\tau(\mathbf{R}_1-\mathbf{r}')\cdot\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_2, W)\right\} \qquad (3.13)$$

At first sight it would seem that there still remain complications due to the long range of the Oseen tensor $\tau$, which is contained in the tensor A. However, wherever this tensor now occurs, either the domain of integration is small [viz. $(4\pi/3)(2a)^3$] or the tensor is multiplied by functions which make the integrand short-ranged, as will be discussed in Section 6. Before we come back in more detail to this last point, we first discuss the case of an unbounded system.

## 4. THE UNBOUNDED SUSPENSION

A popular way to get rid of the complications arising from the presence of walls, i.e., within our formalism, of the terms containing the induced force $\mathbf{f}_0$ on the container walls, is to consider an infinite system *without container walls* right from the start; then $\mathbf{f}_0$ is zero by definition. Furthermore, also $\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_2)$ vanishes, because in an unbounded fluid a single particle subjected to a homogeneous external field of force carries only a force monopole (see, e.g., ref. 1). Equation (3.13) then reduces to

$$6\pi\eta a\mathbf{w}(\mathbf{R}_1) = -\left(1-\frac{11}{2}\phi\right)\mathbf{K} - \frac{3}{16\pi}\phi\int_{|\mathbf{R}_1-\mathbf{R}_2|>2a} d\mathbf{R}_2\,\mathsf{D}(\mathbf{R}_1-\mathbf{R}_2)\cdot\mathbf{K}$$

$$+\frac{3}{4\pi a}\phi\int_{|\mathbf{R}_1-\mathbf{R}_2|>2a} d\mathbf{R}_2\int d\hat{\mathbf{n}}\;\mathsf{A}(\mathbf{R}_1-\mathbf{R}_2-a\hat{\mathbf{n}})\cdot\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_1,\mathbf{R}_2) \qquad (4.1)$$

The last integral in this equation has been evaluated by Batchelor,[6] who combined exact results for the hydrodynamic two-particle problem with numerical methods and found the value $1.55\phi K$, so that

$$6\pi\eta a\mathbf{w}(\mathbf{R}_1) = -\left[1-\left(\frac{11}{2}+1.55\right)\phi\right]\mathbf{K}$$

$$-\frac{3}{16\pi}\phi\int_{|\mathbf{R}_1-\mathbf{R}_2|>2a}\mathsf{D}(\mathbf{R}_1-\mathbf{R}_2)\cdot\mathbf{K} \qquad (4.2)$$

This equation is rather simple in comparison to Eq. (3.13). The price to be paid is the fact that its rhs is not well defined, since the remaining integral does not converge absolutely and its value depends on the way in which the integrations are carried out. For instance, if one first integrates over a spherical region of finite volume and then lets the radius of this region grow to infinity, one obtains

$$\lim_{b \to \infty} \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a \\ R_2 < b}} d\mathbf{R}_2 \, \mathsf{D}(\mathbf{R}_1 - \mathbf{R}_2) \cdot \mathbf{K} = 0 \tag{4.3}$$

while first integrating over a flat slice perpendicular to **K** and then increasing the thickness of the slice to infinity yields

$$\lim_{b \to \infty} \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a \\ |\mathbf{K} \cdot \mathbf{R}_2| < b}} \mathsf{D}(\mathbf{R}_1 - \mathbf{R}_2) \cdot \mathbf{K} = \frac{8\pi}{3} \mathbf{K} \tag{4.4}$$

The question now arises of whether this conditional convergence is an artefact of the unphysical model, the infinite system without walls, or whether it reflects a dependence of **w** on the shape of the container, which would persist even if the latter becomes infinitely large. This problem was noticed by Burgers[3] in 1941, who could, however, no provide a solution. He states that "the possibility that the sedimentation velocity should be dependent upon the shape of the vessel nevertheless does not appear to be readily acceptable." Burgers goes on to mention that during a discussion at the Netherlands Academy of Sciences, "Professor Vening Meinesz raised the question whether it would be possible to solve the problem [i.e., determine the proper sedimentation coefficient] for a suspension extending indefinitely in all directions, provided the boundary condition which in the case of a suspension enclosed in a vessel is imposed by the impermeability of the walls were replaced by an equivalent condition of another type." The argument which Batchelor[6] used to deal with the problem of the conditionally convergent integral may be considered to be in the spirit of Vening Meinesz's remark.

Before we discuss his argument, we derive the following relation for the infinite homogeneous suspension:

$$-\frac{3}{16\pi} \phi \int_{|\mathbf{R}_1 - \mathbf{R}_2| > 2a} d\mathbf{R}_2 \, \mathsf{D}(\mathbf{R}_1 - \mathbf{R}_2) \cdot \mathbf{K} = \pi a^3 \eta \, \Delta \mathbf{V}(\mathbf{R}_1) - \tfrac{1}{2} \phi \mathbf{K} \tag{4.5}$$

In this way the ambiguity of the integral on the lhs is transformed into an ambiguity in the value of the Laplacian of the mean volume velocity. Combination of Eqs. (4.2) and (4.5) yields

$$6\pi\eta a \mathbf{w}(\mathbf{R}_1) = -(1 - 6.55\phi)\mathbf{K} + \pi a^3 \eta \, \Delta \mathbf{V}(\mathbf{R}_1) \tag{4.6}$$

In order to derive the identity (4.5), we first note that the velocity field $\mathbf{v}(\mathbf{r} \mid \mathbf{R})$ which is generated in an unbounded fluid by translational motion of a single sphere with center at $\mathbf{R}$ has the form [which follows also from Eq. (2.13)]

$$\mathbf{v}(\mathbf{r} \mid \mathbf{R}) = \begin{cases} -\mathsf{A}(\mathbf{r}-\mathbf{R}) \cdot \mathbf{K} \dfrac{1}{6\pi\eta a} & \text{for} \quad |\mathbf{r}-\mathbf{R}| > a \\ -\mathbf{K} \dfrac{1}{6\pi\eta a} & \text{for} \quad |\mathbf{r}-\mathbf{R}| < a \end{cases} \tag{4.7}$$

so that, using the formula

$$\Delta \mathsf{T}(\mathbf{r}) = 2\mathsf{D}(\mathbf{r}) \qquad (r > 0) \tag{4.8}$$

one finds

$$\begin{aligned} \Delta \mathbf{v}(\mathbf{r} \mid \mathbf{R}) = &-\Theta(|\mathbf{r}-\mathbf{R}| - a)\, \mathsf{D}(\mathbf{r}-\mathbf{R}) \cdot \mathbf{K} \frac{1}{4\pi\eta} \\ &+ \delta(|\mathbf{r}-\mathbf{R}| - a) \left[ \mathsf{1} - \frac{1}{a^2} (\mathbf{r}-\mathbf{R})^2 \right] \cdot \mathbf{K} \frac{1}{4\pi\eta a^2} \end{aligned} \tag{4.9}$$

To first order in $\phi$ we thus obtain for a homogeneous suspension of spheres, using once more property (3.4),

$$\begin{aligned} \Delta \mathbf{V}(\mathbf{r}) &= \Delta \frac{3}{4\pi a^3} \phi \int d\mathbf{R}\, \mathbf{v}(\mathbf{r} \mid \mathbf{R}) \\ &= \frac{3}{4\pi a^3} \phi \int d\mathbf{R}\, \Delta \mathbf{v}(\mathbf{r} \mid \mathbf{R}) \\ &= -\frac{3}{16\pi^2 \eta a^3} \phi \int_{|\mathbf{r}-\mathbf{R}| > a} \mathsf{D}(\mathbf{r}-\mathbf{R}) \cdot \mathbf{K}\, d\mathbf{R} + \frac{\phi}{2\pi a^3 \eta} \mathbf{K} \end{aligned} \tag{4.10}$$

which is the desired relation (4.5).

It now remains to determine, on general physical grounds, the value $\Delta \mathbf{V}(\mathbf{R}_1)$. One possible chain of arguments runs as follows: There is only one preferential direction in the infinite homogeneous suspension, viz., the direction $\hat{K}$ of the field of force, which we take as the direction of the $z$ axis. Due to symmetry, $V_x$ and $V_y$ must be zero, and $V_z$ can at most depend on $z$. Since, because of incompressibility, the divergence $\partial V_z / \partial z$ of $\mathbf{V}$ vanishes, so does the Laplacian $\Delta \mathbf{V} = (0, 0, \partial^2 V_z / \partial z^2)$. Combination of this result with Eq. (4.6) leads to

$$6\pi\eta a \mathbf{w} = -(1 - 6.55\phi)\mathbf{K} \tag{4.11}$$

Batchelor's argument[6] is essentially equivalent to the one explained above. He states that in the infinite homogeneous suspension the deviatoric part of the stress tensor is uniform, and uses this condition to assign the value $-\frac{1}{2}\phi\mathbf{K}$ to the conditionally convergent integral on the lhs of Eq. (4.5). In Appendix A we show that Batchelor's condition implies $\Delta\mathbf{V}=0$.

We note that the value found in this way for the conditionally convergent integral corresponds to the flat-slice geometry [cf. Eq. (4.4)]. *A posteriori* this is evidently the natural geometry for an infinite system with one preferential direction, and the arguments presented above seem to add very little to that observation. When one accepts that a dependence of the sedimentation velocity $\mathbf{w}$ on the shape of the vessel would be absurd, there can be no doubt that Eq. (4.11) contains *the* correct sedimentation coefficient to order $\phi$. Formally, however, the question of shape dependence has not been settled, for two reasons:

1. In Eq. (3.13), valid for a finite vessel, the first integral on the rhs (which for an unbounded suspension becomes the conditionally convergent integral discussed in this section) clearly does show a dependence on the shape of the vessel. If the sedimentation velocity $\mathbf{w}$ always has the value given in Eq. (4.11), then this shape dependence of the first integral must be compensated by a similar shape dependence of other integrals on the rhs of Eq. (3.13). Such a cancellation is not obvious and needs to be shown.

2. Beenakker and Mazur[7] could be direct calculation determine the mean volume flow $\mathbf{V}(\mathbf{r})$ and the sedimentation velocity $\mathbf{w}(\mathbf{r})$ *in the center*[6] of a spherical container with radius $b$. They found that in the limit of $b$ tending to infinity $\mathbf{w}$ had the value $\mathbf{w}=-(1-6.55\phi)\mathbf{K}$ derived by Batchelor for the infinite system *without walls*. On the other hand, however, they also found that $\mathbf{V}$ is nonzero and finite in the center of the container, even for $b\to\infty$. Therefore, the field $\mathbf{V}(\mathbf{r})$ must contain vortices, since the volume flow is incompressible, a fact which sheads a strange light on the general validity of the symmetry arguments used to determine the value of the conditionally convergent integral in the case of an unbounded suspension. A completely satisfactory understanding of these points can only be achieved by a further analysis of sedimentation in a finite vessel.

## 5. MACROSCOPIC EQUATIONS FOR THE SEDIMENTATION IN A FINITE VESSEL

Returning to Eq. (3.13), we shall find it useful to express the second integral on its rhs in terms of $\Delta\mathbf{V}(\mathbf{R}_1)$, in analogy to the relation (4.5) for

[6] For other points of the container the calculation seemed to present overwhelming difficulties.

the unbounded system. For this purpose we need the following extension of formula (4.8) to all values of $\mathbf{r}$:

$$\Delta\mathsf{T}(\mathbf{r}) = 2\mathsf{D}(\mathbf{r}) + \frac{16\pi}{3}\,\delta(\mathbf{r})\mathsf{1} \tag{5.1}$$

where for integrals containing $\mathsf{D}(\mathbf{r})$ the prescription

$$\int \mathsf{D}(\mathbf{r})\, g(\mathbf{r})\, d\mathbf{r} \equiv \lim_{\varepsilon \downarrow 0} \int_{r > \varepsilon} \mathsf{D}(\mathbf{r})\, g(\mathbf{r})\, d\mathbf{r} \tag{5.2}$$

holds. Combining formulas (5.1) and (3.11), we find

$$\begin{aligned}
\pi\eta a^3\, \Delta\mathbf{V}(\mathbf{R}_1) = {} & -\frac{3}{16\pi}\phi \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > a \\ d(\mathbf{R}_2) > a}} d\mathbf{R}_2\, \mathsf{D}(\mathbf{R}_1 - \mathbf{R}_2)\cdot\mathbf{K} + \frac{\phi}{2}\mathbf{K} \\
& + \frac{N}{\mathscr{V}} \int_{d(\mathbf{R}_2) > a} d\mathbf{R}_2 \left\{ \int d\hat{\mathbf{n}}\, a^2 \frac{a^3}{4} \mathsf{D}(\mathbf{R}_1 - \mathbf{R}_2 - a\hat{\mathbf{n}})\cdot\mathbf{h}_2(\hat{\mathbf{n}} \mid \mathbf{R}_2, W) \right. \\
& \left. + \int ds' \frac{a^3}{4} \mathsf{D}(\mathbf{R}_1 - \mathbf{r}')\cdot\mathbf{f}_0(\mathbf{r}' \mid \mathbf{R}_2, W) \right\} \\
& - \frac{\phi}{2} \int d\hat{\mathbf{n}}\, a^2 \mathbf{h}_2(\hat{\mathbf{n}} \mid \mathbf{R}_1 - a\hat{\mathbf{n}}, W), \qquad d(\mathbf{R}_1) > 2a
\end{aligned} \tag{5.3}$$

Inserting this relation, which generalizes formula (4.5), into Eq. (3.13), we obtain after some straightforward rearrangements of terms

$$\begin{aligned}
& 6\pi\eta a \mathbf{w}(\mathbf{R}_1) \\
& \quad = -(1 - 5\phi)\mathbf{K} + \pi\eta a^3\, \Delta\mathbf{V}(\mathbf{R}_1) \\
& \qquad + \frac{3\phi}{4\pi a^3} \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a \\ d(\mathbf{R}_2) > a}} \int d\hat{\mathbf{n}}\, a^2 \mathsf{A}(\mathbf{R}_1 - \mathbf{R}_2 - a\hat{\mathbf{n}})\cdot\mathbf{h}_2(\hat{\mathbf{n}} \mid \mathbf{R}_1, \mathbf{R}_2) \\
& \qquad + \int_W ds'\, \mathsf{A}(\mathbf{R}_1 - \mathbf{r}')\cdot\mathbf{f}_0(\mathbf{r}' \mid \mathbf{R}_1, W) \\
& \qquad + \frac{N}{\mathscr{V}} \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a \\ d(\mathbf{R}_2) > a}} d\mathbf{R}_2 \left\{ \int d\hat{\mathbf{n}}\, a^2 \mathsf{A}(\mathbf{R}_1 - \mathbf{R}_2 - a\hat{\mathbf{n}})\cdot[\mathbf{h}_2(\hat{\mathbf{n}} \mid \mathbf{R}_1, \mathbf{R}_2, W) \right. \\
& \qquad - \mathbf{h}_2(\hat{\mathbf{n}} \mid \mathbf{R}_1, \mathbf{R}_2) - \mathbf{h}_2(\hat{\mathbf{n}} \mid \mathbf{R}_2, W)] + \int_W ds'\, \mathsf{A}(\mathbf{R}_1 - \mathbf{r}') \\
& \qquad \left. \cdot [\mathbf{f}_0(\mathbf{r}' \mid \mathbf{R}_1, \mathbf{R}_2, W) - \mathbf{f}_0(\mathbf{r}' \mid \mathbf{R}_1, W) - \mathbf{f}_0(\mathbf{r}' \mid \mathbf{R}_2, W)] \right\}
\end{aligned}$$

$$-\frac{N}{\mathscr{V}}\int_{\substack{|\mathbf{R}_1-\mathbf{R}_2|>2a\\ d(\mathbf{R}_2)>a}} d\mathbf{R}_2 \left\{\int d\hat{\mathbf{n}}\, a^2 \mathsf{A}(\mathbf{R}_1-\mathbf{R}_2-a\hat{\mathbf{n}})\cdot\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_2, W)\right.$$

$$\left.+\int_W ds'\,\mathsf{A}(\mathbf{R}_1-\mathbf{r}')\cdot\mathbf{f}_0(\mathbf{r}'\,|\,\mathbf{R}_2, W)\right\}$$

$$+\frac{\phi}{2}\int d\hat{\mathbf{n}}\, a^2\mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{R}_1-a\hat{\mathbf{n}}, W), \qquad d(\mathbf{R}_1)>3a \tag{5.4}$$

The first integral on the rhs of this equation is again the contribution evaluated by Batchelor to be $1.55\phi\mathbf{K}$ (section 4). The remaining integrals represent modifications of the sedimentation velocity due to the container walls. Inspection of these terms suggests that they vanish when the distance between $\mathbf{R}_1$ and the container wall becomes large. That this is indeed so will be shown at greater length in Section 6. We note in passing that the second integral (fourth term) on the rhs of Eq. (5.4) is nothing but the wall correction to the one-particle mobility discussed already by Lorentz and Faxén.[2] Sufficiently far from the walls Eq. (5.4) thus reduces to the form found in Eq. (4.6),

$$6\pi\eta a\mathbf{w}(\mathbf{R}_1) = -(1-6.55\phi)\mathbf{K} + \pi\eta a^3\,\Delta\mathbf{V}(\mathbf{R}_1) \tag{5.5}$$

The value of $\Delta\mathbf{V}(\mathbf{R}_1)$, however, cannot be determined from symmetry arguments, as in Section 4, but must be calculated in a different way.

In principle, one could evaluate the induced forces $\mathbf{f}_0$ and $\mathbf{h}_2$ occurring in Eq. (5.3) and then perform the integrations to find $\Delta\mathbf{V}(\mathbf{R}_1)$. Here, however, we shall follow a different route. The main purpose of Eq. (5.3) was to establish a relation between the quantity $\Delta\mathbf{V}(\mathbf{R}_1)$ and the conditionally convergent integrals involving $\mathsf{D}$, and thus to derive the *local* equation (5.5) for $\mathbf{w}$.

In order to find a closed macroscopic description of sedimentation it is necessary to supplement Eq. (5.5) by a second, manifestly *local* equation for the mean volume flow $\mathbf{V}(\mathbf{r})$. This is easily achieved by averaging Eqs. (2.11) and (2.12), and yields, to linear order in $\phi$,

$$\eta\,\Delta\mathbf{V}(\mathbf{r}) - \nabla P'(\mathbf{r}) = -\frac{3\phi}{4\pi a^3}\int_{d(\mathbf{R}_2)>a} d\mathbf{R}_2\,\mathbf{F}_2(\mathbf{r}\,|\,\mathbf{R}_2, W) \tag{5.6}$$

$$\nabla\cdot\mathbf{V}(\mathbf{r}) = 0 \tag{5.7}$$

where $P'(\mathbf{r})$ is the average of $p'(\mathbf{r})$. Note that Eqs. (5.6) and (5.7) are valid for *all* points inside the container. On the container wall one has stick boundary conditions for $\mathbf{V}(\mathbf{r})$, which follow from the "microscopic" boundary conditions (2.8c) for $\mathbf{v}(\mathbf{r})$,

$$\mathbf{V}(\mathbf{r}) = 0 \qquad \text{for} \quad \mathbf{r}\in W \tag{5.8}$$

It remains to evaluate the inhomogeneous term on the rhs of Eq. (5.6) in terms of the byoyancy-corrected gravitational force $-\mathbf{K}$. With the decomposition (3.6) one has

$$\int_{d(\mathbf{R}_2)>a} d\mathbf{R}_2\, \mathbf{F}_2(\mathbf{r}\,|\,\mathbf{R}_2, W)$$
$$= \int_{d(\mathbf{r}-a\hat{\mathbf{n}})>a} d\hat{\mathbf{n}}\, a^2 \mathbf{f}_2(\hat{\mathbf{n}}\,|\,\mathbf{r}-a\hat{\mathbf{n}}, W)$$
$$= -\int_{d(\mathbf{r}-a\hat{\mathbf{n}})>a} d\hat{\mathbf{n}}\,\frac{1}{4\pi}\mathbf{K} + \int_{d(\mathbf{r}-a\hat{\mathbf{n}})>a} d\hat{\mathbf{n}}\, a^2 \mathbf{h}_2(\hat{\mathbf{n}}\,|\,\mathbf{r}-a\hat{\mathbf{n}}, W) \tag{5.9}$$

The first integral in the last member can easily be evaluated, since the wall may be considered to be flat on the length scale defined by the sphere radius $a$ (see Fig. 1)

$$\int_{d(\mathbf{r}-a\hat{\mathbf{n}})>a} d\hat{\mathbf{n}}\,\frac{1}{4\pi}\mathbf{K} = \begin{cases} \mathbf{K}\, d(\mathbf{r})/2a & \text{for } d(\mathbf{r}) \leqslant 2a \\ \mathbf{K} & \text{for } d(\mathbf{r}) > a \end{cases} \tag{5.10}$$

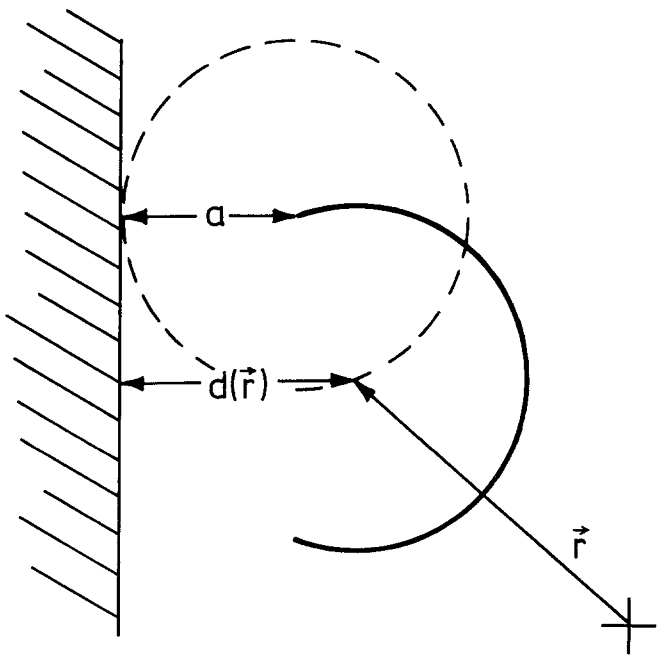


Fig. 1. The sector of integration in Eq. (5.10) is smaller than $4\pi$ for $d(\mathbf{r}) < 2a$.

The reader might wonder why we now insist upon discussing the force term in Eq. (5.8) near the wall, whereas the sedimentation velocity $\mathbf{w}$ was studied only in the case $d(\mathbf{R}_1) \gg a$. The reason is that Eq. (5.5) is an algebraic equation for $\mathbf{w}(\mathbf{r})$, while (5.6) and (5.7) are differential equations, the solution of which at a point $\mathbf{R}_1$ far from the wall will in general also depend on the value of the force term close to the wall.

Evaluation of the second integral in the last member of Eq. (5.9) in terms of $\mathbf{K}$ is nontrivial. For $d(\mathbf{r}) \gg a$ the part $\mathbf{h}_2(\mathbf{r})$ of the induced force is of order $[a/d(\mathbf{r})]^2$ (Section 6). As $\mathbf{r}$ approaches the wall, the integral becomes important. It can be evaluated by combining a multipole expansion of $\mathbf{h}_2$ and a result due to Lorenrz, as was done in ref. 4. This requires extensive calculations, which we hope to present in a future publication. For the time being, we use as a first approximation the monopole contribution (5.10) alone, which will lead to a volume flow $\mathbf{V}$ with qualitatively correct features. The differential equations (5.6) in that case reduces to

$$\eta\, \Delta \mathbf{V}(\mathbf{r}) - \nabla P'(\mathbf{r}) = \frac{3\phi}{4\pi a^3} \left\{ 1 - \Theta(2a - d(\mathbf{r})) \frac{2a - d(\mathbf{r})}{2a} \right\} \mathbf{K} \qquad (5.11)$$

where $\Theta(x)$ denotes the Heaviside function of $x$. The constant force term on the rhs of (5.11) can be included in the pressure; defining

$$P''(\mathbf{r}) \equiv P'(\mathbf{r}) + \frac{3\phi}{4\pi a^3} \mathbf{K} \cdot \mathbf{r} \qquad (5.12)$$

one has

$$\eta\, \Delta \mathbf{V}(\mathbf{r}) - \nabla P''(\mathbf{r}) = \frac{-3\phi}{4\pi a^3} \Theta(2a - d(\mathbf{r})) \frac{2a - d(\mathbf{r})}{2a} \mathbf{K} \qquad (5.13)$$

We now study the solution of this differential equation for several container geometries.

1. First we consider a suspension enclosed by two parallel plates at $x = -b$ and $x = b$ in a Cartesian coordinate system. The $z$ axis is chosen in the direction of the component of $\mathbf{K}$ parallel to the plates. For symmetry reasons there can then be no dependence of the various fields on the $y$ coordinates, and $V_y$ must vanish. Since, moreover, in this case the container is not closed, we have to supplement the boundary condition (5.8) by a condition of no net flow through a cross section,

$$\int_{-b}^{b} V_z \, dx = 0 \qquad (5.14)$$

As one may verify, the solution of Eq. (5.13) with the conditions (5.7), (5.8), and (5.14) is given by

$$V_z = \begin{cases} \frac{3}{2}\phi v_{0z}\left(1-\frac{3}{2}\frac{a}{b}\right)-\frac{9}{2}\phi v_{0z}\left(1-\frac{1}{2}\frac{a}{b}\right)\left(\frac{x}{b}\right)^2 \\ \quad \text{for} \quad |x|<b-2a \\ -\frac{9}{2}\phi v_{0z}\frac{b-|x|}{a}\left[1-2\frac{a}{b}+\left(\frac{a}{b}\right)^2\right] \\ \quad +\frac{9}{4}\phi v_{0z}\left(\frac{b-|x|}{a}\right)^2\left[1-2\left(\frac{a}{b}\right)^2+\left(\frac{a}{b}\right)^3\right]-\frac{3}{8}\phi v_{0z}\left(\frac{b-|x|}{a}\right)^3 \\ \quad \text{for} \quad b-2a<|x|<b \end{cases} \tag{5.15a}$$

$$V_x = 0 \tag{5.15b}$$

$$P'' = \frac{9}{2}\phi v_{0z}\frac{\eta}{a^2}\left[-2\left(\frac{a}{b}\right)^2+\left(\frac{a}{b}\right)^3\right]z + \frac{3\phi}{4\pi a^3}K_x\int_0^x \Theta(|\xi|-b+2a)\frac{|\xi|-b+2a}{2a}\,d\xi \tag{5.16}$$

Here

$$\mathbf{v}_0 = -6\pi\eta a\mathbf{K} \tag{5.17}$$

is the Stokes velocity of a sphere under the influence of an external force $-\mathbf{K}$.

The velocity $V_z$ given by (5.15a) is sketched in Fig. 2. Note that there is a parabolic velocity profile inside the container, varying over distances of the order of $b$, the container dimension. The downward volume flow existing in the center of the container remains even in the limit $b \to \infty$. At $x \approx b/\sqrt{3}$ the volume flow changes sign, and if one extrapolates the parabolic profile to $|x| = b$, one finds an upward velocity

$$(V_z)_{\text{extrapolated}} = -3\phi v_{0z} + O(a/b) \tag{5.18}$$

In reality, the parabolic velocity profile ends at $|x| = b-2a$, and in the following narrow boundary layers the velocity decreases to zero, in agreement with the "microscopic" stick boundary condition. In the bulk, i.e., for $|x| < b-2a$, the Laplacian of the volume flow given by Eq. (5.15) is of order $b^{-2}$, so that one recovers from Eq. (5.5) Batchelor's value for the relative sedimentation velocity in a macroscopic container far from the walls.

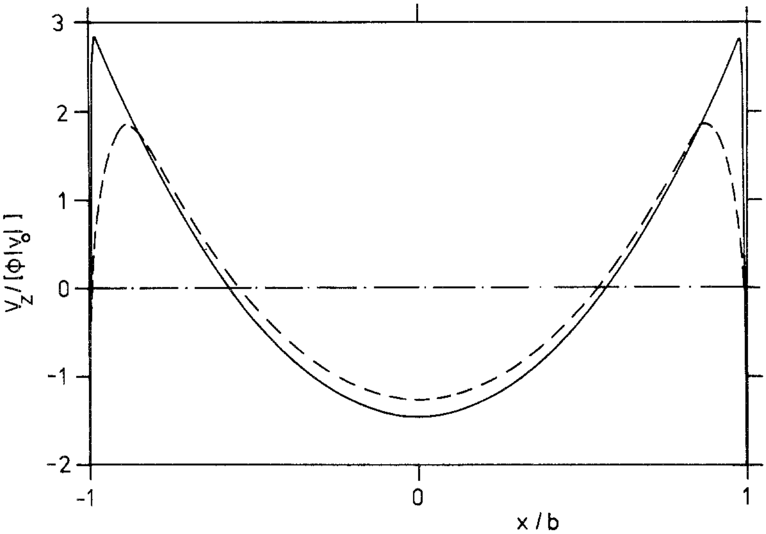


Fig. 2. The mean volume flow due to intrinsic convection between two parallel plates, as given in Eq. (5.15). The dashed line corresponds to $b/a = 10$, the solid line to $b/a = 100$.

2. Let us now consider a suspension enclosed in a spherical container of radius $b$. We choose the origin of the coordinate system in its center. The solution of Eqs. (5.13), (5.7), and (5.8) for this geometry is given by

$$
\mathbf{V}(\mathbf{r}) = \begin{cases}
\frac{9}{2}\phi \mathbf{v}_0 \cdot \Big[ -\frac{\gamma^4}{48}\frac{a}{r}(\mathbf{1} + \hat{\mathbf{r}}^2) - \frac{\gamma^6}{360}\left(\frac{a}{r}\right)^3 (\mathbf{1} - 3\hat{\mathbf{r}}^2) \\
\quad + \left(\frac{\gamma^4}{16\beta} - \frac{\gamma^6}{72\beta^3} + \frac{\beta^3}{144}\right)\mathbf{1} + \frac{1}{48}\left(\frac{r}{a}\right)^3\left(\frac{5}{3}\mathbf{1} - \hat{r}^2\right) \\
\quad + \left(-\frac{\gamma^4}{48\beta^3} + \frac{\gamma^6}{120\beta^5} - \frac{\beta}{48}\right)\left(\frac{r}{a}\right)^2 (21 - \hat{\mathbf{r}}^2)\Big] \\
\quad \text{for} \quad b - 2a < r < b \\
\frac{9}{2}\phi \mathbf{v}_0 \cdot \Big[\left(-\frac{\gamma^4}{48\beta^3} + \frac{\gamma^6}{120\beta^5} + \frac{\gamma}{30} - \frac{\beta}{48}\right)\left(\frac{r}{a}\right)^2 (21 - \hat{\mathbf{r}}^2) \\
\quad + \left(\frac{\gamma^4}{16\beta} - \frac{\gamma^6}{72\beta^3} - \frac{\gamma^3}{18} + \frac{\beta^3}{144}\right)\mathbf{1}\Big] \\
\quad \text{for} \quad r < b - 2a
\end{cases} \tag{5.19}
$$

with

$$\beta \equiv b/a, \qquad \gamma \equiv \beta - 2 \tag{5.20}$$

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For large values of $b/a$ the expression (5.19) reduces to

$$\mathbf{V}(\mathbf{r}) = \begin{cases} \phi \mathbf{v}_0 \cdot (\mathbf{1} - \hat{\mathbf{r}}^2) \left[ -\frac{9}{2}\frac{b-r}{a} + \frac{9}{4}\left(\frac{b-r}{a}\right)^2 - \frac{3}{8}\left(\frac{b-r}{a}\right)^3 \right. \\ \left. \quad + \mathcal{O}\left(\frac{a}{b}\right)\right] \quad \text{for} \quad b-2a<r<b \\ 3\phi \mathbf{v}_0 \cdot \left(\mathbf{1} - \left(\frac{r}{b}\right)^2 (2\mathbf{1} - \hat{\mathbf{r}}) + \mathcal{O}\left(\frac{a}{b}\right)\right] \quad \text{for} \quad r<b-2a \end{cases} \tag{5.21}$$

The bulk solution is similar, and the boundary layer solution identical to those found in the previous case. In the center of the container one finds a downward volume flow

$$\mathbf{V}(\mathbf{r}=0) = 3\phi \mathbf{v}_0 + \mathcal{O}(a/b) \tag{5.22}$$

in agreement with the result obtained by Beenakker and Mazur.[7] If one extrapolates the bulk velocity profile to the container wall, one gets

$$\mathbf{V}_{\text{extrapolated}} = -3\phi(\mathbf{1} - \hat{\mathbf{r}}^2) \cdot \mathbf{v}_0 + \mathcal{O}(a/b) \tag{5.23}$$

in analogy to (5.18). Furthermore, the Laplacian of the volume flow is again of order $b^{-2}$ for $r<b-2a$, so that also in the case of a spherical container the relative sedimentation velocity has Batchelor's value.

We note that the value $3\phi$ on the rhs of Eqs. (5.22) and (5.23) is a consequence of the monopole approximation for the force profile in (5.6). It is to be expected that the influence which higher-order multipoles have on *this* coefficient is comparable to the influence which they have on the first virial coefficient of the sedimentation velocity as calculated by Batchelor, i.e., about 30%.

The convection phenomenon in *homogeneous* suspensions discussed above constitutes an effect of a different nature than the much larger (in magnitude) phenomenon of buoyancy-driven convection due to inhomogeneities in the particle density. It represents a *shape-dependent* effect (in contrast to the *shape-independent* relative sedimentation velocity) which ultimately is caused by the presence of walls. This effect was called essential convection by Beenakker and Mazur[7] and was later named more appropriately intrinsic convection by Nozières.[8]

From the results obtained by explicit solution of Eq. (5.13) one may infer that, as far as the bulk solutions are concerned, the nonuniformity of the force profile in the narrow boundary layer may be neglected if one employs an effective boundary condition

$$\mathbf{V}(\mathbf{r}) = -3\phi(\mathbf{1} - \hat{\mathbf{n}}^2) \cdot \mathbf{v}_0 \qquad (\mathbf{r} \in W) \tag{5.24}$$

for the volume flow, where $\hat{\mathbf{n}}$ here denotes the normal on the wall. As a boundary condition for arbitrary container geometry the relation (5.24) can also be derived by the following argument. The wall of a macroscopic container appears to be flat on the scale of the dimension $a$ of the suspended particles. The behavior of the volume flow close to a wall can therefore be found by solving Eq. (5.13) for a half-space geometry under the condition that this quantity remains finite far away from the wall. This also leads to the relation (5.24).

Very recently Nozières proposed a macroscopic description of the hydrodynamic behavior of a suspension in terms of a set of two *local* coupled equations for the volume flow $\mathbf{V}(\mathbf{r})$ and the relative sedimentation velocity $\mathbf{w}(\mathbf{r})$. Nozières had in mind that the effect of the long-range nature of the hydrodynamic interaction on sedimentation would be taken care of through this coupling of $\mathbf{w}(\mathbf{r})$ and $\mathbf{V}(\mathbf{r})$. His equations, for homogeneous suspensions, read in our notation

$$[\rho_f + \phi(\rho_s - \rho_f)]\mathbf{g} - \nabla P = -\eta\, \Delta \mathbf{V} - \alpha\, \Delta \mathbf{w} \tag{5.25}$$

$$(\rho_s - \rho_f)\mathbf{g} = \lambda \mathbf{w} - \gamma\, \Delta \mathbf{V} \tag{5.26}$$

From Batchelor's result for the sedimentation coefficient[6] and Faxén's theorem,[2] Nozières concludes that

$$\lambda = \frac{9\eta}{2a^2}\,[1 + 6.55\phi + \mathcal{O}(\phi^2)] \tag{5.27}$$

$$\gamma = \frac{3}{4}\,\eta + \mathcal{O}(\phi) \tag{5.28}$$

He also shows that the following Onsager symmetry exists:

$$\alpha = \phi\gamma \tag{5.29}$$

Since there is obviously no volume flow in the limit $\phi \to 0$, it follows that $\Delta\mathbf{V}(\mathbf{r})$ is at least of order $\phi$. Because the coefficient $\alpha$ is proportional to $\phi$, the term $\alpha\Delta\mathbf{w}$ may be omitted from Eq. (5.25) to linear order in the volume fraction. To this order, our Eqs. (5.5) and (5.11) thus agree with those of Nozières. Our derivation of Eqs. (5.5) and (5.11) therefore provides a "microscopic" justification of the latter.[7] In particular, (5.3) and the less general relation (4.5) express, so to say, in rigorous terms the fact that certain long-range hydrodynamic interactions occurring in the sedimentation velocity may indeed be replaced by a local coupling to the volume flow, a fact which, as stated above, formed the basic idea that led Nozières to establish Eqs. (5.25) and (5.26).

[7] In an *infinite* system a justification of Nozières equations has been given very recently by Noetinger,[9] to all orders in the volume fraction, within the framework of the connector formalism developed by Mazur and van Saarloos.[1]

Nozières supplements his equations by a phenomenological boundary condition

$$\mathbf{V} = -\bar{\sigma}\phi(1 - \hat{\mathbf{n}}^2) \cdot \mathbf{w} \tag{5.30}$$

where $\hat{\mathbf{n}}$ again denotes the normal of the wall and "$\mathbf{V}$ and $\mathbf{w}$ are extrapolations of the smooth bulk profiles up to the wall." Nozières admits that his derivation of Eq. (5.30) is questionable, and can only surmise that $\bar{\sigma}$ is of order unity. The "microscopic" theory developed in this paper shows that his boundary condition has the correct form. As Nozières already stated, the value 3 for $\bar{\sigma}$ found above (and to which he concludes by comparison with the result of Beenakker and Mazur) can only be an approximation to lowest order.

## 6. ESTIMATES OF WALL CORRECTION TERMS

In this rather technical section it will be shown that the correction terms to Eq. (5.5), which are due to the presence of container walls, become negligible if one considers a position sufficiently far away from the walls. We shall not try to achieve mathematical rigor. Moreover, only the case of a spherical container of radius $b$ will be treated; we comment on this restriction below.

Let us start by deriving an operator expression for the induced forces, using the tensor $\Pi$. This tensor was introduced in Section 2 as fundamental solution of the stationary equation of motion

$$\nabla \cdot \mathsf{P}'(\mathbf{r}) = \sum_{i=1}^{2} \mathbf{F}_i(\mathbf{r}) \tag{6.1}$$

[cf. Eqs. (2.11) and (2.16)] (for the dilute systems considered it suffices to treat the case that at most two spheres are present). From Eq. (6.1) one finds for the induced surface force density [cf. Eq. (2.9)], using the fact that $\mathsf{P}'$ vanishes inside the spheres,

$$\mathbf{f}_1(\hat{\mathbf{n}}) = \lim_{a' \downarrow a} \hat{\mathbf{n}} \cdot \mathsf{P}'(\mathbf{R}_1 + a'\hat{\mathbf{n}}) \tag{6.2}$$

Combination of this result with formula (2.16) leads to the expression

$$\begin{aligned} \mathbf{f}_1(\hat{\mathbf{n}}) = \lim_{a' \downarrow a} \hat{\mathbf{n}} \cdot \int d\hat{\mathbf{n}}'\, a^2 \Pi(a'\hat{\mathbf{n}} - a\hat{\mathbf{n}}') \cdot \mathbf{f}_1(\hat{\mathbf{n}}') \\ + \hat{\mathbf{n}} \cdot \int d\hat{\mathbf{n}}'\, a^2 \Pi(\mathbf{R}_1 + a\hat{\mathbf{n}} - \mathbf{R}_2 - a\hat{\mathbf{n}}') \cdot \mathbf{f}_2(\hat{\mathbf{n}}') \\ + \hat{\mathbf{n}} \cdot \int d\hat{\mathbf{n}}'\, b^2 \Pi(\mathbf{R}_1 + a\hat{\mathbf{n}} - b\hat{\mathbf{n}}') \cdot \mathbf{f}_0(b\hat{\mathbf{n}}') \end{aligned} \tag{6.3}$$

which can also be written in the short-hand operator notation

$$\mathbf{f}_1 = \mathscr{C}_{11}\mathbf{f}_1 + \mathscr{C}_{12}\mathbf{f}_2 + \mathscr{C}_{10}\mathbf{f}_0 \tag{6.4}$$

The definition of the integral operators $\mathscr{C}_{ij}$ becomes obvious on comparing Eqs. (6.3) and (6.4). When acting on a constant force density, the operator $\mathscr{C}_{11}$ can be further evaluated. Using integral relations for irreducible tensors (see, e.g., ref. 10), one may verify that

$$\hat{\mathbf{r}} \cdot \int \Pi(\mathbf{r} - a\hat{\mathbf{n}})\, d\hat{\mathbf{n}} = \begin{cases} -4\pi\hat{\mathbf{r}} \cdot \Pi(\mathbf{r}) + (a^2/r)\, \mathsf{D}(\mathbf{r}) & \text{for} \quad r > a \\ 0 & \text{for} \quad r < a \end{cases} \tag{6.5}$$

In the limit that $r$ approaches $a$ from above, one obtains from this formula [cf. (2.14)]

$$\lim_{r \downarrow a} \hat{\mathbf{r}} \cdot \int \Pi(\mathbf{r} - a\hat{\mathbf{n}})\, a^2\, d\hat{\mathbf{n}} = 1 \tag{6.6}$$

so that

$$\mathscr{C}_{11}\mathbf{K} = \mathbf{K} \tag{6.7}$$

Using Eq. (6.7) together with the decomposition (3.6) of the induces forces on the spheres, one gets, with $\mathbf{k} \equiv (1/4\pi a^2)\mathbf{K}$,

$$\mathbf{h}_1 = \mathscr{C}_{11}\mathbf{h}_1 + \mathscr{C}_{12}(-\mathbf{k} + \mathbf{h}_2) + \mathscr{C}_{10}\,\mathbf{f}_0 \tag{6.8}$$

A corresponding expression exists of course for $\mathbf{h}_2$,

$$\mathbf{h}_2 = \mathscr{C}_{21}(-\mathbf{k} + \mathbf{h}_1) + \mathscr{C}_{22}\mathbf{h}_2 + \mathscr{C}_{20}\mathbf{f}_0 \tag{6.9}$$

In a way similar to the derivation of Eq. (6.4), one can find an analogous relation for the induced force $\mathbf{f}_0$ on the container wall,

$$\mathbf{f}_0 = \mathscr{C}_{01}(-\mathbf{k} + \mathbf{h}_1) + \mathscr{C}_{02}(-\mathbf{k} + \mathbf{h}_2) + \mathscr{C}_{00}\mathbf{f}_0 \tag{6.10}$$

Here the operators $\mathscr{C}_{00}$ and $\mathscr{C}_{01}$ are defined by

$$(\mathscr{C}_{00}\mathbf{f}_0)(b\hat{\mathbf{n}}) = -\lim_{b' \uparrow b} \hat{\mathbf{n}} \cdot \int \Pi(b'\hat{\mathbf{n}} - b\hat{\mathbf{n}}') \cdot \mathbf{f}_0(b\hat{\mathbf{n}}')\, b^2\, d\hat{\mathbf{n}}' \tag{6.11}$$

$$(\mathscr{C}_{01}\mathbf{f}_1)(b\hat{\mathbf{n}}) = -\hat{\mathbf{n}} \cdot \int \Pi(b\hat{\mathbf{n}} - \mathbf{R}_1 - a\hat{\mathbf{n}}') \cdot \mathbf{f}_1(\hat{\mathbf{n}}')\, a^2\, d\hat{\mathbf{n}}' \tag{6.12}$$

In contrast to $\mathscr{C}_{11}$, the operator $\mathscr{C}_{00}$ maps a constant force density on zero [cf. Eq. (6.5)]

$$\mathscr{C}_{00}\mathbf{k} = 0 \tag{6.13}$$

Note further that $\mathscr{C}_{00}$ does *not* depend on the container radius $b$.

We now first consider the case that there is only one sphere in the container, i.e., $\mathbf{f}_i(\hat{\mathbf{n}}) = \mathbf{f}_i(\hat{\mathbf{n}} \mid \mathbf{R}_i, W)$ $(i = 1, 2)$. The equations (6.8) and (6.10) then reduce to $(i = 1, 2)$

$$-\mathscr{C}_{i0}\mathbf{f}_0 + (1 - \mathscr{C}_{ii})\mathbf{h}_i = 0 \tag{6.14}$$

$$(1 - \mathscr{C}_{00})\mathbf{f}_0 - \mathscr{C}_{0i}\mathbf{h}_i = -\mathscr{C}_{0i}\mathbf{k} \tag{6.15}$$

Introducing the abbreviations $(i = 1, 2)$

$$\mathscr{D} \equiv 1 - \mathscr{C}_{ii} \tag{6.16}$$

$$\mathscr{D}_0 \equiv 1 - \mathscr{C}_{00} \tag{6.17}$$

the solution of the system of equations (6.14), (6.15) reads $(i = 1, 2)$

$$\mathbf{h}_i = -\mathscr{D}^{-1}\mathscr{C}_{i0}(\mathscr{D}_0 - \mathscr{C}_{0i}\mathscr{D}^{-1}\mathscr{C}_{i0})^{-1}\mathscr{C}_{0i}\mathbf{k} \tag{6.18}$$

$$\mathbf{f}_0 = -(\mathscr{D}_0 - \mathscr{C}_{0i}\mathscr{D}^{-1}\mathscr{C}_{i0})^{-1}\mathscr{C}_{0i}\mathbf{k} \tag{6.19}$$

Since $\mathscr{C}_{ii}\mathbf{k} = \mathbf{k}$ $(i = 1, 2)$ [see Eq. (6.7)], it is clear that $\mathscr{D}^{-1}$ only exists on the subspace of the functions on the surface of the spheres which have no monopole moment. But this causes no difficulties in Eqs. (6.18) and (6.19), because the range of the operator $\mathscr{C}_{i0}$ on which $\mathscr{D}^{-1}$ acts consists of functions with vanishing monopole moment:

$$\begin{aligned}
&\int d\hat{\mathbf{n}}\, a^2(\mathscr{C}_{i0}\mathbf{f}_0)(\hat{\mathbf{n}}) \\
&\quad = \int d\hat{\mathbf{n}}\, a^2\hat{\mathbf{n}} \cdot \int d\hat{\mathbf{n}}'\, b^2 \Pi(\mathbf{R}_i + a\hat{\mathbf{n}} - b\hat{\mathbf{n}}') \cdot \mathbf{f}_0(b\hat{\mathbf{n}}') \\
&\quad = \int d\hat{\mathbf{n}}'\, b^2 \int_{r<a} d\mathbf{r}\, \frac{\partial}{\partial \mathbf{r}} \cdot \Pi(\mathbf{R}_i + \mathbf{r} - b\hat{\mathbf{n}}') \cdot \mathbf{f}_0(b\hat{\mathbf{n}}') \\
&\quad = \int d\hat{\mathbf{n}}'\, b^2 \int_{r<a} d\mathbf{r}\, \delta(\mathbf{R}_i + \mathbf{r} - b\hat{\mathbf{n}}') \cdot \mathbf{f}_0(b\hat{\mathbf{n}}') = 0 \qquad (i = 1, 2)
\end{aligned} \tag{6.20}$$

The expressions (6.18), (6.19) can now be used to estimate the magnitude of those wall correction terms on the rhs of Eq. (5.4) in which the induced force corresponds to the case that there is only one sphere in the container. To derive such estimates, we introduce a norm $\|\mathbf{f}\|$ of a surface force density $\mathbf{f}$ by

$$\|\mathbf{f}_i\| \equiv \left[\int d\hat{\mathbf{n}}\, |\mathbf{f}_i(\hat{\mathbf{n}})|^2\right]^{1/2} \qquad (i = 1, 2) \tag{6.21}$$

$$\|\mathbf{f}_0\| \equiv \left[\int d\hat{\mathbf{n}}\, |\mathbf{f}_0(b\hat{\mathbf{n}})|^2\right]^{1/2} \tag{6.22}$$

Making use of the familiar inequalities for norms, one finds

$$\|\mathbf{h}_i(\mathbf{R}_i, W)\| \leqslant \|\mathscr{D}^{-1}\| \; \|\mathscr{C}_{i0}\| \; \|(1-\mathscr{D}_0^{-1}\mathscr{C}_{0i}\mathscr{D}^{-1}\mathscr{C}_{i0})^{-1}\| \times \|\mathscr{D}_0^{-1}\| \; \|\mathscr{C}_{0i}\| \; \|\mathbf{k}\| \qquad (i=1,2) \tag{6.23}$$

$$\|\mathbf{f}_0(\mathbf{R}_i, W)\| \leqslant \|(1-\mathscr{D}_0^{-1}\mathscr{C}_{0i}\mathscr{D}^{-1}\mathscr{C}_{i0})^{-1}\| \; \|\mathscr{D}_0^{-1}\| \; \|\mathscr{C}_{0i}\| \; \|\mathbf{k}\| \tag{6.24}$$

In Appendix B we show that

$$\|\mathscr{C}_{0i}\| < \text{const} \cdot \frac{1}{b(b-R_i-a)} \qquad (i=1,2) \tag{6.25}$$

$$\|\mathscr{C}_{i0}\| < \text{const} \cdot \frac{1}{b-R_i-a} \qquad (i=1,2) \tag{6.26}$$

We note in passing that these upper bounds on the norms of the operators $\mathscr{C}_{i0}$ and $\mathscr{C}_{0i}$ become useless when sphere $i$ touches the container wall, i.e., for $R_i+a=b$. Some estimations needed later, on the other hand, would be greatly faciliated if one knew that $\mathscr{C}_{i0}$ and $\mathscr{C}_{0i}$ where always bounded. Fortunately, we may indeed suppose this in the present context for the following reason. It is quite clear that the divergence of the bounds (6.25), (6.26) has nothing to do with the properties of the hydrodynamic interactions over large distances, which give rise to the question of shape dependence discussed in this paper, but rather stems from the singularity of the tensor $\Pi(\mathbf{r})$ for $r \to 0$. Without losing the essential features of our system we may therefore assume that the *statistical* properties of the spheres are those of a gas of hard spheres of radius $a+\delta$, with some positive constant $\delta$ very small compared to $a$, while the *hydrodynamically relevant* radius of the spheres remains equal to $a$. Such a system, for which the bounds (6.25), (6.26) are always finite, is not a worse model of a real suspension than the original one (i.e., that with $\delta=0$).

We furthermore assume that the operators $\mathscr{D}_0^{-1}$ and $\mathscr{D}^{-1}$ are bounded. We did not succeed in proving this property, although it is physically evident: After all, boundedness of, e.g., $\mathscr{D}_0^{-1}$ only means that the magnitude of the force density induced on the container by an incoming pressure tensor field is at most of the order of magnitude of this incoming field.

Using these assumptions, one finds from Eqs. (6.23) and (6.24) with the aid of (6.25) and (6.26) (for sufficiently large $b$) the estimates ($i=1,2$)

$$\|\mathbf{h}_i(\mathbf{R}_i, W)\| < \text{const} \cdot \frac{1}{b(b-R_i-a)} \tag{6.27}$$

$$\|\mathbf{f}_0(\mathbf{R}_i, W)\| < \text{const} \cdot \frac{1}{b(b-R_i-a)} \tag{6.28}$$

From relation (6.28) one sees that the wall correction to the single-particle mobility, the second integral of the right member of Eq. (5.4), vanishes as $1/b$ if $b$ tends to infinity, while $R_1/b$ remains bounded by a given constant smaller than unity,

$$\begin{aligned}
&\left|\int d\hat{\mathbf{n}}\, b^2 \mathsf{A}(\mathbf{R}_1 - b\hat{\mathbf{n}}) \cdot \mathbf{f}_0(b\hat{\mathbf{n}} \,|\, \mathbf{R}_1, W)\right| \\
&\quad \leqslant b^2 \frac{\text{const}}{b - R_1} \int d\hat{\mathbf{n}}\, |\mathbf{f}_0(b\hat{\mathbf{n}} \,|\, \mathbf{R}_1, W)| \\
&\quad \leqslant b^2 \frac{\text{const}}{b - R_1} \left( \int d\hat{\mathbf{n}}\, |\mathbf{f}_0(b\hat{\mathbf{n}} \,|\, \mathbf{R}_1, W)|^2 \right)^{1/2} \left( \int d\hat{\mathbf{n}} \right)^{1/2} \\
&\quad \leqslant b^2 \frac{\text{const}}{b - R_1} \|\mathbf{f}_0(\mathbf{R}_1, W)\| \\
&\quad \leqslant \text{const} \cdot \frac{b}{(b - R_1)(b - R_1 - a)} = \mathcal{O}\left(\frac{1}{b}\right)
\end{aligned} \tag{6.29}$$

Here use was made of the Schwarz inequality.

The norm $\|\mathbf{f}_i\|$ defined in Eq. (6.21) of course does not in general tell us anything about the absolute value of the function $\mathbf{f}_i$ at a particular value of its argument. If the function is sufficiently smooth, however, the norm (6.12) provides a correct order-of-magnitude estimate of the maximum of $|\mathbf{f}_i(\hat{\mathbf{n}})|$ with respect to $\hat{\mathbf{n}}$. With this in mind one can convince oneself that the last two terms in Eq. (5.4) also vanish as $1/b$ if the container radius $b$ tends to infinity while $R_1/b$ stays bounded by a constant smaller than unity. Indeed, the induced force $\mathbf{h}_2(\hat{\mathbf{n}} \,|\, \mathbf{R}_2, W)$ in these terms is a smooth function of $\hat{\mathbf{n}}$ and $\mathbf{R}_2$, since $\mathbf{R}_2$ is restricted to the neighborhood of $\mathbf{R}_1$ and thus far away from the container wall which generates the force density $\mathbf{h}_2(\hat{\mathbf{n}} \,|\, \mathbf{R}_2, W)$. Furthermore, as $b$ (and with it the distance between $\mathbf{R}_1$ and the container wall) grows, this force density can only become smoother still.

In order to treat the remaining wall correction terms on the rhs of Eq. (5.4), it is necessary to solve the ful system of equations (6.8)–(6.10) with both particles present. For the estimation of these terms it turns out to be handy to insert in Eqs. (6.8)–(6.10) in front of $\mathbf{h}_1$ and $\mathbf{h}_2$ the projector $\mathscr{H}$ given by

$$(\mathscr{H}\mathbf{f})(\hat{\mathbf{n}}) \equiv \mathbf{f}(\hat{\mathbf{n}}) - \frac{1}{4\pi} \int d\hat{\mathbf{n}}'\, \mathbf{f}(\hat{\mathbf{n}}') \tag{6.30}$$

which annihilates the monopole part of a force density $\mathbf{f}$ but leaves higher-order multipoles unchanged. The reason for inserting the projector $\mathscr{H}$ lies in the fact that we can derive for the operator $\mathscr{C}_{0i}\mathscr{H}$ $(i=1, 2)$ the upper bound (cf. Appendix B)

$$\|\mathscr{C}_{0i}\mathscr{H}\| < \frac{\text{const}}{b(b-R_i-a)^2} \qquad (i=1, 2) \tag{6.31}$$

which is better than (6.25) for $b-R_i \gg a$.

Introducing the abbreviations

$$\mathscr{E}_i \equiv (1-\mathscr{D}^{-1}\mathscr{C}_{ij}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{ji}\mathscr{H})^{-1} \qquad (i, j=1, 2) \tag{6.32}$$

and

$$\begin{aligned}\mathscr{G} \equiv \{1 &- \mathscr{D}_0^{-1}\mathscr{C}_{01}\mathscr{H}\mathscr{E}_1\mathscr{D}^{-1}(\mathscr{C}_{10}+\mathscr{C}_{12}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{20}) \\ &- \mathscr{D}_0^{-1}\mathscr{C}_{02}\mathscr{H}\mathscr{E}_2\mathscr{D}^{-1}(\mathscr{C}_{20}+\mathscr{C}_{21}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{10})\}^{-1}\mathscr{D}_0^{-1}\end{aligned} \tag{6.33}$$

one finds for

$$\delta\mathbf{f}_0(\mathbf{R}_1, \mathbf{R}_2, W) \equiv \mathbf{f}_0(\mathbf{R}_1, \mathbf{R}_2, W) - \mathbf{f}_0(\mathbf{R}_1, W) - \mathbf{f}_0(\mathbf{R}_2, W) \tag{6.34}$$

from Eqs. (6.8)–(6.10) the formula

$$\begin{aligned}\delta\mathbf{f}_0 = &-\mathscr{G}[\mathscr{C}_{01}\mathscr{H}(\mathscr{E}_1-1)\mathscr{D}^{-1}\mathscr{C}_{10}+\mathscr{C}_{01}\mathscr{H}\mathscr{E}_1\mathscr{D}^{-1}\mathscr{C}_{12}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{20} \\ &+\mathscr{C}_{02}\mathscr{H}\mathscr{E}_2\mathscr{D}^{-1}(\mathscr{C}_{20}+\mathscr{C}_{21}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{10})] \\ &\times(1-\mathscr{D}_0^{-1}\mathscr{C}_{01}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{10})^{-1}\mathscr{D}_0^{-1}\mathscr{C}_{01}\mathbf{k} \\ &-\mathscr{G}[\mathscr{C}_{02}\mathscr{H}(\mathscr{E}_2-1)\mathscr{D}^{-1}\mathscr{C}_{20}+\mathscr{C}_{02}\mathscr{H}\mathscr{E}_2\mathscr{D}^{-1}\mathscr{C}_{21}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{10} \\ &+\mathscr{C}_{01}\mathscr{H}\mathscr{E}_1\mathscr{D}^{-1}(\mathscr{C}_{10}+\mathscr{C}_{12}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{20})] \\ &\times(1-\mathscr{D}_0^{-1}\mathscr{C}_{02}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{20})^{-1}\mathscr{D}_0^{-1}\mathscr{C}_{02}\mathbf{k} \\ &-\mathscr{G}\mathscr{C}_{01}\mathscr{H}\mathscr{E}_1\mathscr{D}^{-1}\mathscr{C}_{12}(1+\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{21})\mathbf{k} \\ &-\mathscr{G}\mathscr{C}_{02}\mathscr{H}\mathscr{E}_2\mathscr{D}^{-1}\mathscr{C}_{21}(1+\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{12})\mathbf{k}\end{aligned} \tag{6.35}$$

We collect those terms on the rhs of this lengthy formula which are more easy to estimate in a function $\psi$ defined by

$$\delta\mathbf{f}_0 = \psi - \mathscr{D}_0^{-1}\mathscr{C}_{02}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{20}\mathscr{D}_0^{-1}\mathscr{C}_{01}\,\mathbf{k} - \mathscr{D}_0^{-1}\mathscr{C}_{02}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{21}\,\mathbf{k} \tag{6.36}$$

Using (6.25), (6.26), and (6.31) as well as the relation (cf. Appendix B)

$$\|\mathscr{C}_{12}\| < \frac{\text{const}}{|\mathbf{R}_1-\mathbf{R}_2|\,(|\mathbf{R}_1-\mathbf{R}_2|-2a)} \tag{6.37}$$

one may verify that

$$\|\psi\| < \text{const} \cdot [\|\mathscr{C}_{01}\mathscr{H}\| \; \|\mathscr{C}_{12}\| + \|\mathscr{C}_{01}\mathscr{H}\| \; \|\mathscr{C}_{02}\| + \|\mathscr{C}_{02}\mathscr{H}\| \; \|\mathscr{C}_{12}\| \, (\mathscr{C}_{02}\| + \|\mathscr{C}_{12}\|)] \tag{6.38}$$

holds, and that, furthermore,

$$\int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a+\delta \\ R_2 < b-a-\delta}} d\mathbf{R}_2 \, \|\mathscr{C}_{12}\| < \text{const} \cdot b \tag{6.39}$$

$$\int_{R_2 < b-a-\delta} d\mathbf{R}_2 \, \|\mathscr{C}_{02}\| < \text{const} \cdot b \log \frac{b}{\delta} \tag{6.40}$$

$$\int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a+\delta \\ R_2 < b-a-\delta}} d\mathbf{R}_2 \, \|\mathscr{C}_{02}\mathscr{H}\| \; \|\mathscr{C}_{12}\| \, (\|\mathscr{C}_{02}\| + \|\mathscr{C}_{12}\|) < \text{const} \cdot \frac{1}{b^2} \tag{6.41}$$

Here it was once more assumed that $b$ is sufficiently large and that the ratio $R_1/b$ is bounded by a constant smaller than unity.

We are now in a position to treat the contribution to the wall corrections in Eq. (5.4) that contains $\psi$. Applying again Schwarz's inequality, one has

$$\begin{aligned} &\left| \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a+\delta \\ R_2 < b-a-\delta}} \int d\hat{\mathbf{n}} \, b^2 \mathsf{A}(\mathbf{R}_1 - b\hat{\mathbf{n}}) \cdot \psi(b\hat{\mathbf{n}}) \right| \\ &\quad \leqslant \text{const} \cdot \frac{b^2}{b - R_1} \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a+\delta \\ R_2 < b-a-\delta}} \|\psi\| \\ &\quad \leqslant \text{const} \cdot \frac{b^2}{b-R_1} \left[ \frac{b}{b(b-R_1)^2} + \frac{1}{(b-R_1)^2} \log \frac{b}{\delta} + \frac{1}{b^2} \right] \\ &\quad = \mathcal{O}\left( \frac{1}{b} \log b \right) \to 0 \qquad (b \to \infty) \end{aligned} \tag{6.42}$$

The wall corrections containing $\delta \mathbf{f}_0 - \psi$ require more careful study. Using only the function norm defined in Eqs. (6.21) and (6.22) and the estimates for the various operators with respect to this norm would not suffice to show that those terms also vanish as $b \to \infty$. Instead, we need to introduce a second function norm by

$$\|\mathbf{f}_0\|_1 \equiv \int |\mathbf{f}_0(b\hat{\mathbf{n}})| \; d\hat{\mathbf{n}} \tag{6.43}$$

because with respect to this norm we can give the estimate (cf. Appendix B)

$$\|\mathscr{C}_{02}\|_1 < \text{const} \cdot \frac{1}{bR_2} \log \frac{b}{\delta} \qquad (R_2 < b - a - \delta) \tag{6.44}$$

which is a factor $(\log b)/b$ better than (6.25) and (6.31) for $R_2 \approx b$.[8] Using (6.44) together with (6.25) and (6.26), one finds for $b$ sufficiently large and $R_1/b$ bounded by a constant $< 1$

$$\begin{aligned}
&\left| \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a+\delta \\ R_2 < b-a-\delta}} \int d\hat{\mathbf{n}}\, b^2 \mathsf{A}(\mathbf{R}_1 - b\hat{\mathbf{n}})(\mathscr{D}_0^{-1}\mathscr{C}_{02}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{20}\mathscr{D}_0^{-1}C_{01}\,\mathbf{k})(b\hat{\mathbf{n}}) \right| \\
&\quad \leqslant \text{const} \cdot \frac{b^2}{b - R_1} \int_{R_2 < b-a-\delta} d\mathbf{R}_2 \frac{\log b}{bR_2} \int d\hat{\mathbf{n}}\, |(\mathscr{D}^{-1}\mathscr{C}_{20}\mathscr{D}_0^{-1}\mathscr{C}_{01}\mathbf{k})(\hat{\mathbf{n}})| \\
&\quad \leqslant \text{const} \cdot \log b \int_{R_2 < b-a-\delta} d\mathbf{R}_2 \frac{1}{R_2} \left\{ \int d\hat{\mathbf{n}}\, |(\mathscr{D}^{-1}\mathscr{C}_{20}\mathscr{D}_0^{-1}\mathscr{C}_{01}\mathbf{k})(\hat{\mathbf{n}})|^2 \right\}^{1/2} \\
&\quad \leqslant \text{const} \cdot \log b \int_0^{b-a-\delta} dR_2\, R_2 \|\mathscr{C}_{20}\|\, \|\mathscr{C}_{01}\| \\
&\quad \leqslant \text{const} \cdot \log b \int_0^{b-a-\delta} dR_2\, R_2 \frac{1}{b - R_2 - a} \frac{1}{b(b - R_1 - a)} \\
&\quad \leqslant \text{const} \cdot \frac{\log b}{b} \int_0^{b-a-\delta} dR_2 \frac{1}{b - R_2 - a} \\
&\quad = \mathscr{O}\left[ \frac{1}{b} (\log b)^2 \right] \to 0 \qquad (b \to \infty)
\end{aligned} \tag{6.45}$$

Finally, the wall correction containing the last term of the rhs of Eq. (6.36) can be estimated as follows:

$$\begin{aligned}
&\left| \int_{\substack{R_2 < b-a-\delta \\ |\mathbf{R}_1 - \mathbf{R}_2| > 2a+\delta}} d\mathbf{R}_2 \int d\hat{\mathbf{n}}\, b^2 \mathsf{A}(\mathbf{R}_1 - b\hat{\mathbf{n}})(\mathscr{D}_0^{-1}\mathscr{C}_{02}\mathscr{H}\mathscr{D}^{-1}\mathscr{C}_{21}\mathbf{k})(b\hat{\mathbf{n}}) \right| \\
&\quad \leqslant \text{const} \cdot \frac{b^2}{b - R_1} \int_{b - a^{1/3}(b-R_1)^{2/3} < R_2 < b-a-\delta} \|\mathscr{C}_{02}\|_1\, \|\mathscr{C}_{21}\| \\
&\quad\quad + \text{const} \cdot \frac{b^2}{b - R_1} \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a+\delta \\ b - a^{1/3}(b-R_1)^{2/3} > R_2}} d\mathbf{R}_2\, \|\mathscr{C}_{02}\mathscr{H}\|\, \|\mathscr{C}_{21}\|
\end{aligned}$$

[8] We cannot work exclusively with this $L^1$ norm since in other configurations and for different operators one obtains poorer estimates than with the norm introduced before.

$$\leqslant \text{const} \cdot \log b \int_{b - a^{1/3}(b - R_1)^{2/3} < R_2 < b - a - \delta} \frac{1}{R_2} \frac{1}{|\mathbf{R}_1 - \mathbf{R}_2|^2}$$

$$+ \text{const} \cdot \int_{R_2 < b - a^{1/3}(b - R_1)^{2/3}} d\mathbf{R}_2 \frac{1}{(b - R_2)^2} \frac{1}{|\mathbf{R}_1 - \mathbf{R}_2|^2}$$

$$\leqslant \text{const} \cdot \log \mathrm{b} \int_{b - a^{1/3}(b - R_1)^{2/3}}^{b - a - \delta} dR_2 \frac{1}{R_2} \frac{R_2}{R_1} \log \frac{1 + R_1/R_2}{1 - R_1/R_2}$$

$$+ \text{const} \cdot \frac{1}{a^{2/3}(b - R_1)^{4/3}} \int_{|\mathbf{R}_1 - \mathbf{R}_2| < b + R_1} \frac{1}{|\mathbf{R}_1 - \mathbf{R}_2|^2}$$

$$\leqslant \text{const} \cdot \log b \log \frac{b}{b - a^{1/3}(b - R_1)^{2/3}} + \text{const} \cdot \frac{R_1 + b}{(b - R_1)^{4/3}}$$

$$= \mathcal{O}\left(\frac{\log b}{b^{1/3}}\right) \to 0 \qquad (b \to \infty) \tag{6.46}$$

Collecting the results (6.42), (6.45), and (6.46), we can write, for large values of $b$,

$$\left| \int_{\substack{|\mathbf{R}_1 - \mathbf{R}_2| > 2a + \delta \\ R_2 < b - a - \delta}} d\mathbf{R}_2 \int d\hat{\mathbf{n}}\, b^2 \mathsf{A}(\mathbf{R}_1 - b\hat{\mathbf{n}}) \cdot \delta \mathbf{f}_0(b\,\hat{\mathbf{n}} \,|\, \mathbf{R}_1, \mathbf{R}_2, W) \right|$$

$$= \mathcal{O}\left(\frac{\log b}{b^{1/3}}\right) \tag{6.47}$$

This wall correction thus also bcomes small for large vessels.

The only term on the rhs of Eq. (5.4) remaining to be studied is the one containing

$$\mathbf{h}_2(\mathbf{R}_1, \mathbf{R}_2, W) - \mathbf{h}_2(\mathbf{R}_1, \mathbf{R}_2) - \mathbf{h}_2(\mathbf{R}_2, W) \tag{6.48}$$

Since the estimation of this term is lengthy but easier than that of the term containing $\delta \mathbf{f}_0$, we shall not give it explicitly here. One finds a better bound than that in (6.47).

Note that in the above estimates $R_1$ was allowed to grow proportionally to $b$ as the latter increases. We have therefore shown that even if $b - R_1 \ll b$ (though $b - R_1 \gg a$), the wall corrections vanish for $b \to \infty$. In this sense it was demonstrated that the validity of Eq. (5.5) is not restricted to positions near the center of the container if the latter is large enough.

To conclude this section, we discuss the various assumptions that we had to make.

The fact that we restricted ourselves to the case of spherical container does not appear to be an important limitation, since we did not make use of the spherical symmetry. It should not be an essential problem to extend our result to, e.g., the case of a general convex container, although the additional technical effort would probably be considerable.

At one point we had to use the fact that the force density $\mathbf{h}_2(\hat{\mathbf{n}} \mid \mathbf{R}_2, W)$ is a smooth function. The arguments used there are somewhat "handwaving," but they can undoubtedly be formalized if one considers it necessary.

Finally, we had to assume that $\mathscr{D}_0^{-1}$ and $\mathscr{D}^{-1}$ are bounded operators. We find this assumption physically very plausible; still, it would be nice if it someone could give a mathematical proof.

## 7. CONCLUDING REMARKS

The theory proposed in this paper combines the simplicity of Nozières' phenomenological theory of the coupling between sedimentation and convection with the more fundamental character of the approach based on the treatment of many-body hydrodynamic interactions, which has been explored in Leiden during recent years. All the essential results of Nozières, who himself characterized his paper as "largely speculative," have been confirmed, at least to first order in the volume fraction and for homogeneous suspensions.

The surprising phenomenon of intrinsic convection may therefore by now be considered well established from a theoretical point of view. The experimental situation is less clear. The only experimental reference we know of in which a possibly intrinsic convection flow was reported is a nearly 40-year-old paper by Kinosita.[11] More recent experiments conducted by Buscall *et al.*,[12] on the other hand, seem to compare favorably with Noetinger's[9] calculation of the sedimentation velocity assuming *zero volume flow*.[9] This does not, however, necessarily indicate the absence of intrinsic convection, because, in contrast to Kinosita, Buscall *et al.* just measured the downward movement of the meniscus separating the suspension from the pure solvent. It will be only slightly distorted by intrinsic convection, which tends to lift the meniscus near the walls and to lower it in the center of the vessel, since the lateral inhomogeneity thus generated gives rise to an additional convective current leveling the meniscus again. We expect this additional convection to be negligible far away from the

[9] In a sharp corner of a vessel the effective boundary condition (5.30) would be incompatible with the incompressibility of the volume flow if it were to hold *exactly at the wall.* Misinterpreting the character of the effective boundary condition, Noetinger concluded that for a parallelepipedic container the coefficient $\bar{\sigma}$, and consequently the intrinsic convection flow, should vanish.

meniscus, where the intrinsic convection should behave qualitatively as in the examples investigated in Section 5. We hope that experimentalists will feel challenged to provide the first unambiguous observation of intrinsic convection.

On the theoretical side interesting problems remain, too. In the first place, we refer here to the need for a better calculation of the coefficient $\tilde{\sigma}$ in the effective boundary condition (5.28), which has up to now only be evaluated in monopole aproximation. Furthermore, it would be nice if one could improve on the rather coarse estimate of the wall correction terms given in Section 6. The physical expectation is that these terms are of order $a/d(R_1)$, which is much stronger than what we were able to show.

## APPENDIX A

We show in this Appendix that Batchelor's condition that the average of the deviatoric part d of the stress tensor is uniform implies that the Laplacian of the average volume flow vanishes.

For the case of one sphere with center at $\mathbf{R}$ immersed in an infinite fluid, the tensor $\mathsf{d}(\mathbf{r})$ only depends on $\mathbf{r}-\mathbf{R}$ (for simplicity we take the sphere elastically isotropic). In the fluid part

$$\mathsf{d}(\mathbf{r}) = 2\eta \left[\frac{\partial}{\partial \mathbf{r}} \mathbf{v}(\mathbf{r}\,|\,\mathbf{R})\right]^s, \qquad |\mathbf{r}-\mathbf{R}| > a \tag{A.1}$$

holds, with $\mathbf{v}(\mathbf{r}\,|\,\mathrm{R})$ given by formula (4.7). In the solid part one has

$$\mathsf{d}(\mathbf{r}) = \mathsf{d}_s(\mathbf{r}-\mathbf{R}), \qquad |\mathbf{r}-\mathbf{R}| < a \tag{A.2}$$

where $\mathsf{d}_s$ depends on the elastic properties of the sphere but does not need to be specified here.

The average of d then is, to first order in the volume fraction, given by

$$\begin{aligned}\langle \mathsf{d}(\mathbf{r})\rangle &= \frac{N}{\mathscr{V}}\left\{\int_{|\mathbf{r}-\mathbf{R}|>a} d\mathbf{R}\; 2\eta \left[\frac{\partial}{\partial \mathbf{r}} \mathbf{v}(\mathbf{r}\,|\,\mathbf{R})\right]^s + \int_{|\mathbf{r}-\mathbf{R}|<a} d\mathbf{R}\;\; \mathsf{d}_s(\mathbf{r}-\mathbf{R})\right\} \\ &= \frac{N}{\mathscr{V}}\left\{2\eta \int d\mathbf{R} \left[\frac{\partial}{\partial \mathbf{r}} \mathbf{v}(\mathbf{r}\,|\,\mathbf{R})\right]^s + \int_{r'<a} d\mathbf{r}'\; \mathsf{d}_s(\mathbf{r}')\right\} \\ &= 2\eta \left[\frac{\partial}{\partial \mathbf{r}} \mathbf{V}(\mathbf{r})\right]^s + \frac{N}{\mathscr{V}} \int_{r'<a} d\mathbf{r}'\; \mathsf{d}(\mathbf{r}')\end{aligned} \tag{A.3}$$

Here we used the fact that $(\partial/\partial \mathbf{r})\, \mathbf{v}(\mathbf{r}\,|\,\mathbf{R})$ vanishes for $|\mathbf{r}-\mathbf{R}| < a$ [cf.

Eq. (4.7)]. From Batchelor's condition that $\langle \mathbf{d}(\mathbf{r}) \rangle$ is uniform, one thus finds

$$0 = \frac{\partial}{\partial \mathbf{r}} \cdot \langle \mathbf{d}(\mathbf{r}) \rangle = 2\eta \, \Delta \mathbf{V}(\mathbf{r}) \tag{A.4}$$

## APPENDIX B

In this Appendix we derive various bounds for operators needed in Section 6. We start with the operator $\mathscr{C}_{01}$. For an arbitrary function $\mathbf{f}(\hat{\mathbf{n}})$ one finds, using Schwatz's inequality,

$$\begin{aligned}
&|(\mathscr{C}_{01}\mathbf{f})(b\hat{\mathbf{n}})|^2 \\
&= \left| \hat{\mathbf{n}} \cdot \int \Pi(b\hat{\mathbf{n}} - \mathbf{R}_1 - a\hat{\mathbf{n}}') \cdot \mathbf{f}(\mathbf{n}') \, a^2 \, d\hat{\mathbf{n}}' \right|^2 \\
&\leqslant a^4 \int d\hat{\mathbf{n}}' \, \hat{\mathbf{n}} \cdot \Pi(b\hat{\mathbf{n}} - \mathbf{R}_1 - a\hat{\mathbf{n}}') : \Pi(b\hat{\mathbf{n}} - \mathbf{R}_1 - a\hat{\mathbf{n}}') \cdot \hat{\mathbf{n}} \int d\hat{\mathbf{n}}'' \, |f(\hat{\mathbf{n}}'')|^2 \\
&\leqslant a^4 \int d\hat{\mathbf{n}}' \frac{1}{|b\hat{\mathbf{n}} - \mathbf{R}_1 - a\hat{\mathbf{n}}'|^4} \, \|\mathbf{f}\|^2 \\
&= \frac{4\pi a^4}{(|b\hat{\mathbf{n}} - \mathbf{R}_1|^2 - a^2)^2} \, \|\mathbf{f}\|^2
\end{aligned} \tag{B.1}$$

For $\|\mathscr{C}_{01}\mathbf{f}\|$ one obtains with the aid of this formula

$$\begin{aligned}
|\mathscr{C}_{01}\mathbf{f}\| &= \left[ \int d\mathbf{n} \, |(\mathscr{C}_{01} f)(b\hat{\mathbf{n}})|^2 \right]^{1/2} \\
&\leqslant \left( \int d\hat{\mathbf{n}} \frac{4\pi a^4}{(|b\hat{\mathbf{n}} - \mathbf{R}_1|^2 - a^2)^2} \right)^{1/2} \|\mathbf{f}\| \\
&= \frac{4\pi a^2}{[(b - R_1)^2 - a^2]^{1/2} [(b + R_1)^2 - a^2]^{1/2}} \, \|\mathbf{f}\| \\
&< \frac{4\pi a^2}{b(b - R_1 - a)} \, \|\mathbf{f}\|
\end{aligned} \tag{B.2}$$

which proves the relation (6.25).

For $\mathscr{C}_{01}\mathscr{H}$, a better bound than (6.25) can be given in the case $b-R_1 \gg a$. Since the integral of $\mathscr{H}\mathbf{f}$ over the surface of the unit sphere vanishes, one may write

$$
\begin{aligned}
&(\mathscr{C}_{01}\mathscr{H}\mathbf{f})(b\hat{\mathbf{n}}) \\
&\quad = -\hat{\mathbf{n}}\cdot\int \Pi(\hat{\mathbf{n}}b-\mathbf{R}_1-a\hat{\mathbf{n}}')\cdot(\mathscr{H}\mathbf{f})(\hat{\mathbf{n}}')\,a^2\,d\hat{\mathbf{n}}' \\
&\quad = -\hat{\mathbf{n}}\cdot\int \left[a^2\Pi(\hat{\mathbf{n}}b-\mathbf{R}_1-a\hat{\mathbf{n}}')-a^2\Pi(\hat{\mathbf{n}}b-\mathbf{R}_1)\right]\cdot(\mathscr{H}\mathbf{f})(\hat{\mathbf{n}}')\,d\hat{\mathbf{n}}'
\end{aligned}
\tag{B.3}
$$

If $b-R_1 \gg a$, the elements of the tensor between square brackets in the last member of Eq. (B.3) are of order $(a/|b(\hat{\mathbf{n}}-\mathbf{R}_1|)^3$. Therefore one has

$$
\begin{aligned}
\|\mathscr{C}_{01}\mathscr{H}\mathbf{f}\| &\leqslant \text{const}\cdot\left(\int d\hat{\mathbf{n}}\,\frac{a^6}{|b\hat{\mathbf{n}}-\mathbf{R}_1|^6}\right)^{1/2}\|\mathscr{H}\mathbf{f}\| \\
&\leqslant \text{const}\cdot\frac{a^3}{(b+R_1)(b-R_1)^2}\,\|\mathscr{H}\mathbf{f}\|, \qquad b-R_1\gg a
\end{aligned}
\tag{B.4}
$$

Since different multipoles are orthogonal with respect to each other, $\|\mathscr{H}\mathbf{f}\|$ is smaller than or equal to $\|\mathbf{f}\|$. The restriction $b-R_1\gg a$ in relation (B.4) can be omitted if one replaces the factor $b-R_1$ in the last member of this expression by $b-R_1-a$ [cf. Eq. (B.2)]. Thus, the bound (6.31) is obtained.

The derivations of the bounds (6.26) for $\|\mathscr{C}_{10}\|$ and (6.37) for $\|\mathscr{C}_{12}\|$ proceed analogously to the derivation of the relation (6.25) for $\|\mathscr{C}_{01}\|$:

$$
\begin{aligned}
\|\mathscr{C}_{10}\mathbf{f}_0\| &= \left[\int d\hat{\mathbf{n}}\left|\int b^2\,d\hat{\mathbf{n}}'\,\hat{\mathbf{n}}\cdot\Pi(\mathbf{R}_1+a\hat{\mathbf{n}}-b\hat{\mathbf{n}}')\cdot\mathbf{f}_0(b\hat{\mathbf{n}}')\right|^2\right]^{1/2} \\
&\leqslant b\left(\int d\hat{\mathbf{n}}\int d\hat{\mathbf{n}}'\,\frac{1}{|\mathbf{R}_1+a\hat{\mathbf{n}}-b\mathbf{n}'|^4}\right)^{1/2}\|\mathbf{f}_0\| \\
&= \frac{4\pi b}{[(b+R_1+a)(b+R_1-a)(b-R_1+a)(b-R_1-a)]^{1/2}}\,\|\mathbf{f}_0\| \\
&\leqslant \text{const.}\,\frac{1}{b-a-R_1}\,\|\mathbf{f}_0\|
\end{aligned}
\tag{B.5}
$$

$$
\begin{aligned}
\|\mathscr{C}_{12}\mathbf{f}\| &= \left[\int d\hat{\mathbf{n}}\left|\int a^2\,d\hat{\mathbf{n}}'\,\hat{\mathbf{n}}\cdot\Pi(\mathbf{R}_1+a\hat{\mathbf{n}}-\mathbf{R}_2-a\hat{\mathbf{n}}')\cdot\mathbf{f}(\hat{\mathbf{n}}')\right|^2\right]^{1/2} \\
&\leqslant a^2\left(\int d\hat{\mathbf{n}}\int d\hat{\mathbf{n}}'\,\frac{1}{|\mathbf{R}_1-\mathbf{R}_2+a\hat{\mathbf{n}}-a\hat{\mathbf{n}}'|^4}\right)^{1/2}\|\mathbf{f}\| \\
&= \frac{4\pi a^2}{|\mathbf{R}_1-\mathbf{R}_2|\,(|\mathbf{R}_1-\mathbf{R}_2|^2-4a^2)^{1/2}}\,\|\mathbf{f}\| \\
&\leqslant \text{const}\cdot\frac{1}{|\mathbf{R}_1-\mathbf{R}_2|\,(|\mathbf{R}_1-\mathbf{R}_2|-2a)}\,\|\mathbf{f}\|
\end{aligned}
\tag{B.6}
$$

Finally, the bound (6.44) for $\mathscr{C}_{02}$ with respect to the norm $\|\cdot\|_1$ defined in Eq. (6.43) can also be found in a straightforward manner:

$$\begin{aligned}\|\mathscr{C}_{02}\mathbf{f}\|_1 &= \int d\hat{\mathbf{n}} \left| \int a^2\, d\hat{\mathbf{n}}'\, \hat{\mathbf{n}} \cdot \Pi(b\hat{\mathbf{n}} - \mathbf{R}_2 - a\hat{\mathbf{n}}') \cdot \mathbf{f}(\hat{\mathbf{n}}') \right| \\ &\leqslant \int d\hat{\mathbf{n}} \frac{a^2}{(|b\hat{\mathbf{n}} - \mathbf{R}_2| - a)^2} \int d\hat{\mathbf{n}}'\, |\mathbf{f}(\hat{\mathbf{n}}')| \\ &= \frac{2\pi a^2}{R_2 b} \left( \log \frac{b + R_2 - a}{b - R_2 - a} - \frac{1}{b + R_2 - a} + \frac{1}{b - R_2 - a} \right) \|\mathbf{f}\|_1 \\ &< \text{const} \cdot \frac{a^2}{R_2 b} \log \frac{b}{\delta} \end{aligned} \tag{B.7}$$

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