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Stresses in inhomogeneous suspensions

Jan K. G. Dhont
Forschungszentrum Jülich, IFF/Weiche Materie, D-52425 Jülich, Germany

W. J. Briels
Computational Dispersion Rheology, Department of Applied Physics, University of Twente, Postbus 217, 7500 AE Enschede, The Netherlands

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A microscopic expression for the divergence of the stress tensor is derived for inhomogeneous suspensions of rigid colloidal particles of arbitrary shape. This expression is valid for arbitrary large gradients in the shear rate, concentration, and orientational order parameters. The corresponding Navier–Stokes equation is a necessary ingredient to describe phenomena like shear-banding, phase separation kinetics, and phase coexistence under shear flow conditions. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1495842]

I. INTRODUCTION

There are a number of nonequilibrium phenomena in systems subjected to shear flow, where strong inhomogeneities play an important role, both in transient kinetics and in patterned stationary states. Examples of such phenomena are phase separation and phase coexistence under shear flow conditions, and shear-banding. In order to describe such phenomena, a closed set of equations of motion must be derived for the suspension flow velocity, for the density and relevant order parameters. These equations of motion should account for the occurrence of gradients in the shear rate, concentration, and order parameters. Since within interfacial regions these gradients can be very large, it is probably not sufficient to use truncated gradient expansions of the equations of motion. Instead, the equations of motion should account for all higher order gradient contributions in closed form. The equation of motion for the suspension flow velocity is the Navier–Stokes equation, for which the divergence of the suspension stress tensor must be specified. *In the present paper we will derive an expression for the divergence of the suspension stress tensor which includes contributions due to, possibly very large, gradients in shear rate, concentration, and orientational order parameters*.

A special feature of simple shear flow is, that it constitutes a nonconservative external field. One cannot define a Hamiltonian for a system subjected to simple shear flow, and therefore, one can not define a chemical potential μ. Furthermore, even if a chemical potential could be defined locally, mass transport would only be proportional to $\nabla\mu$, when gradients are small and the system is close to equilibrium. It is not known how good the approximation is to ignore these problems, and to assume that mass transport is driven by linear gradients in some local chemical potential. We therefore chose to develop a microscopic approach to derive equations of motion, which does not rely on thermodynamic arguments. As a first step toward the derivation of a complete set of equations of motion, we shall derive a microscopic expression for the divergence of the suspension stress tensor simply by adding forces that act on an infinitesimally small volume element.

The expression for the divergence of the stress tensor derived in the present paper is of a simplicity that allows for its explicit evaluation in terms of shear rate, concentration, and the relevant order parameters. In a subsequent paper[1] we shall perform this explicit evaluation for uniaxial, long and thin, rigid hard-core rods. As will turn out, equations of motion contain convolution-type of integrals. Gradient expanding these convolution-type integrals up to fourth order gradients leads to equations of motion that are similar (but not identical) in structure as the commonly used equations of motion that are obtained from the above mentioned thermodynamic considerations. For systems with large gradients, such a truncation may not be valid anymore, and the convolution-type integrals should be kept as they stand.

The suspension flow velocity $\mathbf{U}(\mathbf{r},t)$ at position $\mathbf{r}$ and time t can be found by solving the Navier–Stokes equation,

$$\rho_m\left[\frac{\partial \mathbf{U}(\mathbf{r},t)}{\partial t}+\mathbf{U}(\mathbf{r},t)\cdot\nabla\mathbf{U}(\mathbf{r},t)\right]=\nabla\cdot\boldsymbol{\Sigma}(\mathbf{r},t), \tag{1}$$

where ρ_m is the mass-density of the suspension. The suspension flow will be assumed incompressible, that is,

$$\nabla\cdot\mathbf{U}(\mathbf{r},t)=0. \tag{2}$$

To apply these equations in order to describe the above mentioned phenomena, the divergence of the suspension stress tensor must be expressed in terms of shear rate, concentration, and orientational order parameters, including stress contributions arising from gradients in these quantities.

A recent paper by Fang *et al.*[7] discusses mass transport driven by gradients in the stress. Here, gradients are assumed to be small enough that their explicit contributions to the stress tensor need not be taken into account. Perhaps the present theory may help to quantify how small spatial gradients should actually be in order to be able to use constitutive equations for homogeneous systems to describe mass transport.

The only work we are aware of that deals with stresses in inhomogeneous colloidal suspensions is due to Prosperetti and co-workers.[2–4] In these studies, ensemble averaged quantities as considered by Lhuillier[5,6] are computed numerically for inhomogeneous suspensions of *spherical colloidal particles*. Here, we shall derive a closed, general expression for the divergence of the suspension stress tensor which is valid for *arbitrarily shaped, rigid colloidal particles*. The main result of our calculation extends the expression for the stress tensor as derived by Batchelor[8] and Doi and Edwards[9] to inhomogeneous suspensions. In fact, the derivation presented here considerably simplifies the original derivation as described by these authors, even though our result is valid also for strongly inhomogeneous systems.

This paper is organized as follows: Sec. II outlines the general idea of our derivation. Sections III–V deal with the calculation of the various stress contributions. In Sec. VI it is shown from force balance on the Smoluchowski time scale, that two terms cancel, leading to a relatively simple expression for the divergence of the total stress tensor, that is valid for arbitrarily large gradients in shear rate, concentration, and relevant order parameters.

II. THE BASIC IDEA

For most colloidal systems, the suspension flow velocity $\mathbf{U}(\mathbf{r},t)$ at some point $\mathbf{r}$ and time t is simply equal to the ensemble average of the microscopic velocity $\mathbf{v}$ (Refs. 2–6) (the conditions under which this relation holds are discussed in Appendix A),

$$\mathbf{U}(\mathbf{r},t)=\langle \mathbf{v}(\mathbf{r}|\boldsymbol{\Gamma})\rangle, \tag{3}$$

where Γ is used as a short-hand notation for the phase space variables of the colloidal particles. For an assembly of N rigid, uniaxial rods, for example, Γ is the $6N$ dimensional vector comprised of N center-of-mass coordinates and N unit vectors that characterize the orientations of the rods. Note that $\mathbf{v}(\mathbf{r}|\boldsymbol{\Gamma})$ is either the local solvent velocity at $\mathbf{r}$ (when $\mathbf{r}$ at time t is within the solvent), or is the velocity of a volume element within the core of a colloidal particle. The ensemble average $\langle \cdots \rangle$ is taken with respect to the probability density function of the phase space variables Γ, that complies with a chosen ensemble. In the present analysis there is no need to actually specify the ensemble.

Let $P(\boldsymbol{\Gamma},t)$ denote the probability density function of $\boldsymbol{\Gamma}$ that complies with the chosen ensemble. By definition we then have

$$\mathbf{U}(\mathbf{r},t)=\langle \mathbf{v}(\mathbf{r})\rangle=\int d\boldsymbol{\Gamma}\, P(\boldsymbol{\Gamma},t)\, \mathbf{v}(\mathbf{r}|\boldsymbol{\Gamma}). \tag{4}$$

In the derivation of the general expression for the divergence of the stress tensor, we shall encounter the ensemble average,

$$\langle \nabla^2 \mathbf{v}(\mathbf{r})\rangle=\int d\boldsymbol{\Gamma}\, P(\boldsymbol{\Gamma},t)\, \nabla^2 \mathbf{v}(\mathbf{r}|\boldsymbol{\Gamma}).$$

Since the Laplace operator can be taken in front of the phase-space integral, and the suspension flow velocity is given by Eq. (4), it trivially follows that

$$\langle \nabla^2 \mathbf{v}(\mathbf{r})\rangle=\nabla^2 \mathbf{U}(\mathbf{r},t). \tag{5}$$

This result will be of importance later in this paper.

Similarly to the definition (3) of the suspension velocity, the divergence of the suspension stress tensor is simply the ensemble averaged force per unit volume that acts at a given point in the dispersion. We shall simply add up all forces due to interactions between solvent molecules and colloidal particles in order to obtain the divergence of the suspension stress tensor.

Consider a rectangular volume element δV located at $\mathbf{r}$, with linear dimensions δ_x, δ_y, and δ_z in the x-, y-, and z-direction, respectively. In the formal limit that the size of the volume element vanishes, the ensemble averaged total force per unit volume of the surrounding material on the volume element is nothing but the divergence of the stress tensor that should be used in the Navier–Stokes equation (1). This force consists of three parts: forces that arise from interactions between colloidal particles outside on those within the rectangular volume element, from interactions between solvent molecules and colloidal particles, and from interactions between solvent molecules on either side of the boundary of the volume element. The corresponding stress tensors will be referred to as the "particle–particle stress tensor" $\boldsymbol{\Sigma}^{pp}$, the "particle–solvent stress tensor" $\boldsymbol{\Sigma}^{ps}$, and the "solvent–solvent stress tensor" $\boldsymbol{\Sigma}^{ss}$, respectively. The total stress tensor is simply the sum of these stress contributions,

$$\nabla\cdot\boldsymbol{\Sigma}=\nabla\cdot\boldsymbol{\Sigma}^{pp}+\nabla\cdot\boldsymbol{\Sigma}^{ps}+\nabla\cdot\boldsymbol{\Sigma}^{ss}. \tag{6}$$

These three contributions will be considered separately in Secs. III–V, respectively.

The position coordinate of colloidal particle j will be denoted as $\mathbf{r}_j$. Its orientational state will be denoted as ω_j. For uniaxial, rigid rods, one would have,

$$\omega_j=\hat{\mathbf{u}}_j, \quad \text{uniaxial rods},$$

where $\hat{\mathbf{u}}_j$ is the unit vector in the direction of the long axis of the rod. For biaxial rigid bodies, the variable ω_j stands for two perpendicular unit vectors (or, equivalently, three angles) that are needed to specify the orientation.

We should emphasize at this point, that Eq. (3) is not generally valid, also for other situations where, for example, sedimentation is considered. For the more general case of two-phase flow, where one of the components is not of a colloidal nature, the conditions for the validity of Eq. (3) that are given in Appendix A are generally not satisfied. In the present analysis, that applies to suspensions of rigid colloidal particles, the validity of Eq. (3) is only needed for the evaluation of the solvent–solvent contribution to the stress tensor [see Sec. V, Eq. (31) in particular].

III. THE PARTICLE–PARTICLE STRESS TENSOR $\boldsymbol{\Sigma}^{pp}$

The force that colloidal particles outside the volume element exert on those within the volume element is equal to

$$\sum_j{}^{\star}\ \mathbf{F}_j,$$

where $\mathbf{F}_j$ is the force on colloidal particle j, and the $\star$ on the summation is used to indicate that the summation ranges only over those colloidal particles that are inside the volume element, that is, for which $\mathbf{r}_j\in\delta V$. It is used here that mutual interactions between colloidal particles within the volume element can not give rise to a net force on that volume element. The force per unit volume, for formally vanishing size of the volume element, is thus equal to

$$\nabla\cdot\boldsymbol{\Sigma}^{pp}=\lim_{\delta_x,\delta_y,\delta_z\to 0}\frac{1}{\delta_x\,\delta_y\,\delta_z}\sum_{j=1}^{N}\langle\chi_{\mathbf{r}}(\mathbf{r}_j)\,\mathbf{F}_j\rangle, \tag{7}$$

where N is the total number of colloidal particles in the system under consideration, $\mathbf{r}_j$ is the position coordinate of a colloidal particle, and $\chi_{\mathbf{r}}$ is the characteristic function of the rectangular volume element that was introduced in the previous section. The characteristic function is defined as

$$\begin{aligned}\chi_{\mathbf{r}}(\mathbf{R})&=1\quad\text{when}\quad\mathbf{R}\in\delta V,\\&=0\quad\text{otherwise}.\end{aligned}\tag{8}$$

The subscript "$\mathbf{r}$" on the characteristic function is used to indicate that the volume element δV is located at position $\mathbf{r}$. The characteristic function in Eq. (7) effectively limits the summation to colloidal particles that are inside δV, that is, for which $\mathbf{r}_j\in\delta V$. Furthermore, the total force $\mathbf{F}_j$ on the jth colloidal particle due to interactions with all other colloidal particles is equal to[10,11]

$$\mathbf{F}_j=-\nabla_j\Psi-k_BT\nabla_j\ln\{P\}, \tag{9}$$

where Ψ is the total potential energy of the assembly of N rods in the suspension, and P is the probability density function of the phase space coordinates of all the colloidal particles; $-\nabla_j\Psi$ is the force due to potential interactions, and $-k_BT\nabla_j\ln\{P\}$ is the Brownian force, where ∇_j is the gradient operator with respect to $\mathbf{r}_j$. Since [with $\delta(\mathbf{r}-\mathbf{r}_j)$ the three-dimensional delta distribution],

$$\lim_{\delta_x,\delta_y,\delta_z\to 0}\chi_{\mathbf{r}}(\mathbf{r}_j)/\delta_x\,\delta_y\,\delta_z=\delta(\mathbf{r}-\mathbf{r}_j), \tag{10}$$

as is easily verified by integration of both sides with respect to $\mathbf{r}_j$, Eq. (7) immediately leads to

$$\nabla\cdot\boldsymbol{\Sigma}^{pp}=\sum_{j=1}^{N}\langle\delta(\mathbf{r}-\mathbf{r}_j)\,\mathbf{F}_j\rangle. \tag{11}$$

Together with Eq. (9) for the forces, this is the microscopic expression for the contribution to the divergence of the stress tensor which is due to intercolloidal particle forces. This particle–particle contribution to the stress tensor contains the osmotic pressure contribution to the total pressure of the suspension. The Brownian contribution in Eq. (9) for the force on a colloidal particle gives rise to the ideal gas osmotic pressure, while the interaction contribution accounts for the effect of interactions on the osmotic pressure.

An alternative, intuitively may be more appealing expression, can be obtained by introducing the conditional probability density function (pdf) for the phase space coordinates,

$$\{\mathbf{r}_1,\ldots,\mathbf{r}_{j-1},\mathbf{r}_{j+1},\ldots,\mathbf{r}_N,\omega_1,\ldots,\omega_N\},$$

with a prescribed position coordinate $\mathbf{r}_j$ of colloidal particle j. This conditional pdf is equal to

$$P^{(c)}=P/P(\mathbf{r}_j,t), \tag{12}$$

where P is the pdf for all the phase space coordinates (including $\mathbf{r}_j$), and $P(\mathbf{r}_j,t)$ is the 1-particle pdf for $\mathbf{r}_j$. The latter pdf is related to the local number density ρ of colloidal particles as

$$P(\mathbf{r},t)=\frac{1}{N}\rho(\mathbf{r},t).$$

Denoting the ensemble average with respect to $P^{(c)}$ as $\langle\cdots\rangle^{(c)}$, Eq. (11) can be written, for identical colloidal particles, as

$$\nabla\cdot\boldsymbol{\Sigma}^{pp}=\rho(\mathbf{r},t)\,\langle\mathbf{F}\rangle^{(c)}(\mathbf{r},t). \tag{13}$$

Here, $\langle\mathbf{F}\rangle^{(c)}(\mathbf{r},t)$ is the conditionally averaged force on a colloidal particle that resides at $\mathbf{r}$ at time t. Since $\nabla\cdot\boldsymbol{\Sigma}$ is the body force per unit volume on the suspension, this expression could almost have been written down immediately; the right-hand side of Eq. (13) is indeed the force on a single colloidal particle, given that its center is in $\mathbf{r}$, multiplied by the number of particles per unit volume at that point. The above reasoning assumes mono-disperse suspensions [where the conditional pdf in Eq. (12) is independent of the particle number index j], but is easily extended to the polydisperse case.

It is shown in Appendix B that for the calculation of the suspension shear viscosity of a *homogeneous system*, Eq. (11) for the stress tensor reduces to

$$\boldsymbol{\Sigma}^{pp}=\sum_{j=1}^{N}\langle\delta(\mathbf{r}-\mathbf{r}_j)\,\mathbf{r}_j\,\mathbf{F}_j\rangle. \tag{14}$$

This is identical to the expression for the particle–particle stress tensor as derived (for the first time) by Batchelor,[8] except that the prefactor $1/V$ (with V the volume of the system) is now replaced by the delta distribution $\delta(\mathbf{r}-\mathbf{r}_j)$. In fact, volume averaging of the above expression by acting with the operator $(1/V)\int d\mathbf{r}(\cdots)$ on both sides, reproduces Batchelor's expression for the particle–particle stress contribution,

$$\overline{\boldsymbol{\Sigma}}^{pp}=\frac{1}{V}\sum_{j=1}^{N}\langle\mathbf{r}_j\,\mathbf{F}_j\rangle, \tag{15}$$

where the overbar is used to indicate volume averaging. Note that, after such a volume averaging, this stress contribution becomes position independent, so that one might conclude that its divergence vanishes. This would imply that this stress contribution is irrelevant, since it does not contribute to the Navier–Stokes equation. The traditional way to interpret the volume avaraged contribution to the stress is as follows. In an explicit evaluation of the right-hand side of Eq. (15), one finds that it is proportional to $\boldsymbol{\Gamma}+\boldsymbol{\Gamma}^T$, with $\boldsymbol{\Gamma}$ the velocity gradient tensor. The velocity gradient tensor is then interpreted as $(\nabla\mathbf{U})^T$, with $\mathbf{U}$ the suspension velocity. The contribution to the Navier–Stokes equation is then $\sim\nabla^2\mathbf{U}$, assuming incompressible suspensions, and the proportionality constant contributes to the suspension viscosity. In this way, a nonzero contribution to the suspension viscosity of the particle–particle stress tensor is found from Eq. (15). This interpretation is explicitly accounted for by the delta distribution in Eq. (14).

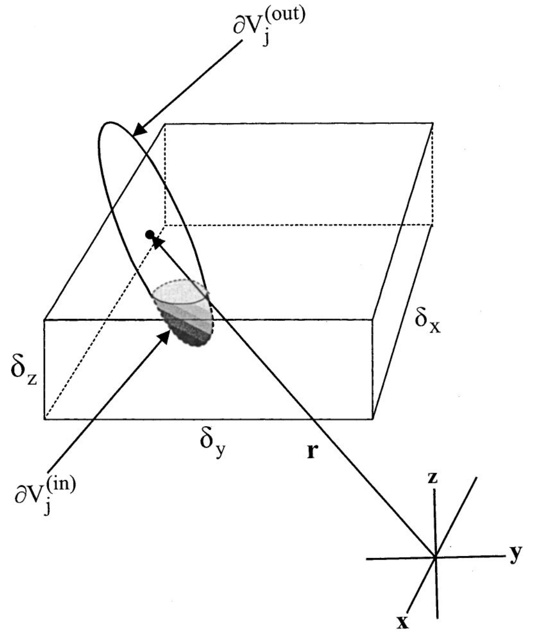

FIG. 1. The rectangular volume element δV at position $\mathbf{r}$, intersects with the core of colloidal particle j with its position coordinate $\mathbf{r}_j$ outside the volume element. $\partial V_j^{(\text{out})}$ is that part of the surface area ∂V_j of the colloidal particle that is outside the volume element, $\partial V_j^{(\text{in})}$ is the part inside.

IV. THE PARTICLE–SOLVENT STRESS TENSOR Σ^{ps}

The particle–solvent stress arises from forces on the volume element due to interactions between colloidal particles and solvent molecules. These forces are mediated to the volume element by colloidal particles that intersect with the surface enclosing the rectangular volume element that was introduced in Sec. II. Consider first the force that is mediated to the solvent within the volume element by a colloidal particle with its position coordinate outside the volume element (see Fig. 1). The instantaneous force that the colloidal particle exerts on the solvent inside the volume element is equal to

$$-\int_{\partial V_j^{(\text{in})}(\mathbf{r}_j,\omega_j)} d\mathbf{S}' \cdot \boldsymbol{\sigma}(\mathbf{r}'). \tag{16}$$

Here, the surface area $\partial V_j^{(\text{in})}$ is that part of the surface area of the colloidal particle that is inside the volume element (see Fig. 1). This range of integration depends both on the position $\mathbf{r}_j$ of colloidal particle j and its orientational state, which is denoted by ω_j, as discussed in Sec. II. Furthermore, $d\mathbf{S}'$ is the normal surface element on the surface area of the colloidal particle, and $\boldsymbol{\sigma}$ is the microscopic stress tensor of the solvent. The minus sign in Eq. (16) arises from the fact that $d\mathbf{S}' \cdot \boldsymbol{\sigma}(\mathbf{r}')$ is equal to $dS'\,\mathbf{f}^h(\mathbf{r}')$, with $\mathbf{f}^h(\mathbf{r}')$ the force per unit area that the fluid exerts on the surface element $d\mathbf{S}'$, which is minus the force that this surface element exerts on the fluid. In terms of this hydrodynamic force, Eq. (16) is more conveniently written as

$$-\int_{\partial V_j^{(\text{in})}(\mathbf{r}_j,\omega_j)} dS'\,\mathbf{f}^h(\mathbf{r}').$$

The ensemble averaged force $\mathbf{F}^{\text{out}}$ of all colloidal particles outside the volume element on the solvent inside the element, is thus equal to,

$$\mathbf{F}^{\text{out}} = -\left\langle \sum_{j=1}^{N} [1-\chi_{\mathbf{r}}(\mathbf{r}_j)] \oint_{\partial V_j(\mathbf{r}_j,\omega_j)} dS'\,\mathbf{f}^h(\mathbf{r}')\,\chi_{\mathbf{r}}(\mathbf{r}') \right\rangle, \tag{17}$$

where, as before, $\chi_{\mathbf{r}}$ is the characteristic function for the volume element. The characteristic function $1-\chi_{\mathbf{r}}(\mathbf{r}_j)$ for the volume outside the volume element assures that in the summation over all colloidal particles, only those are counted which are outside the volume element. Furthermore, the characteristic function $\chi_{\mathbf{r}}(\mathbf{r}')$ assures that only points $\mathbf{r}'$ on the surface of the colloidal particle which are inside the volume element are taken into account. Including the characteristic function in the integrand in Eq. (17) allows for the extension of the integration range to the entire surface area $\partial V_j(\mathbf{r}_j,\omega_j)$ of the jth colloidal particle.

Similarly, in case a colloidal particle is located inside the volume element, that is, when $\mathbf{r}_j \in \delta V$, the instantaneous force that the colloidal particle exerts on the solvent outside the volume element is equal to

$$-\int_{\partial V_j^{(\text{out})}(\mathbf{r}_j,\omega_j)} dS'\,\mathbf{f}^h(\mathbf{r}'),$$

with $\partial V_j^{(\text{out})}$ the part of the surface area of the colloidal particle located outside the volume element (see Fig. 1). This is minus the force that is exerted on the colloidal particle by the solvent outside the volume element. Hence, similarly as before, the ensemble averaged force $\mathbf{F}^{\text{in}}$ on the volume element due to interactions between solvent molecules outside and colloidal particles inside the volume element is found to be equal to

$$\mathbf{F}^{\text{in}} = \left\langle \sum_{j=1}^{N} \chi_{\mathbf{r}}(\mathbf{r}_j) \oint_{\partial V_j(\mathbf{r}_j,\omega_j)} dS'\,\mathbf{f}^h(\mathbf{r}')\,[1-\chi_{\mathbf{r}}(\mathbf{r}')] \right\rangle, \tag{18}$$

where it is used again that $1-\chi_{\mathbf{r}}(\mathbf{r}')$ is the characteristic function for the volume outside the volume element.

From the representation (10) of the delta distribution it is thus found that

$$\begin{aligned}\nabla \cdot \boldsymbol{\Sigma}^{ps} &= \lim_{\delta_x,\delta_y,\delta_z \to 0} [\mathbf{F}^{\text{out}} + \mathbf{F}^{\text{in}}]/\delta_x\,\delta_y\,\delta_z \\ &= \sum_{j=1}^{N} \langle \delta(\mathbf{r}-\mathbf{r}_j)\,\mathbf{F}_j^h \rangle \\ &\quad - \sum_{j=1}^{N} \left\langle \oint_{\partial V_j(\mathbf{r}_j,\omega_j)} dS'\,\delta(\mathbf{r}-\mathbf{r}')\,\mathbf{f}^h(\mathbf{r}') \right\rangle,\end{aligned} \tag{19}$$

where

$$\mathbf{F}_j^h = \oint_{\partial V_j(\mathbf{r}_j,\omega_j)} dS'\,\mathbf{f}^h(\mathbf{r}') \tag{20}$$

is the total force that the solvent exerts on the jth colloidal particle.

It is shown in Appendix C, that for a *homogeneous system*, Eq. (19) leads to the following expression for the particle–solvent stress tensor:

$$\boldsymbol{\Sigma}^{ps}=\sum_{j=1}^{N}\left\langle \delta(\mathbf{r}-\mathbf{r}_j)\oint_{\partial V_j(\mathbf{r}_j,\omega_j)} d\mathbf{S}'\,(\mathbf{r}'-\mathbf{r}_j)\,\mathbf{f}^h(\mathbf{r}')\right\rangle . \tag{21}$$

Upon volume averaging this yields

$$\bar{\boldsymbol{\Sigma}}^{ps}=\frac{1}{V}\sum_{j=1}^{N}\left\langle \oint_{\partial V_j(\mathbf{r}_j,\omega_j)} d\mathbf{S}'\,(\mathbf{r}'-\mathbf{r}_j)\,\mathbf{f}^h(\mathbf{r}')\right\rangle , \tag{22}$$

which reproduces Batchelor's expression for the particle–solvent stress tensor.[8] The interpretation of the difference between the two expressions (21) and (22) is the same as for the particle–particle stress tensor, as discussed at the end of the previous section.

V. THE SOLVENT–SOLVENT STRESS TENSOR $\boldsymbol{\Sigma}^{ss}$

The force per unit volume that the solvent outside the volume element δV exerts on the solvent inside, for formally vanishing size of the volume element, is equal to

$$\nabla\cdot\boldsymbol{\Sigma}^{ss}=\lim_{\delta_x,\delta_y,\delta_z\to 0}\frac{1}{\delta_x\,\delta_y\,\delta_z}\left\langle\int_{A_s} d\mathbf{S}'\cdot\sigma(\mathbf{r}')\right\rangle , \tag{23}$$

where A_s is the part of the surface area of the volume element that is occupied by solvent, which is the surface area of the volume element minus the part that is cut by cores of colloidal particles (see Fig. 2). Here, $d\mathbf{S}'$ points outward of δV. The subscript "s" on the integration A_s refers to "solvent." For an incompressible solvent we have

$$\sigma(\mathbf{r}')=\eta_0[\nabla'\mathbf{v}(\mathbf{r}')+(\nabla'\mathbf{v}(\mathbf{r}'))^T]-p(\mathbf{r}')\,\hat{\mathbf{I}}, \tag{24}$$

with η_0 the solvent shear viscosity and $\mathbf{v}$ the solvent flow velocity. Furthermore, p is the mechanical pressure in the solvent, and $\hat{\mathbf{I}}$ is the identity tensor. The superscript "T" stands for the transpose of a tensor. Note that since $\nabla^2 p(\mathbf{r})=0$ within the incompressible solvent, p is entirely determined by the boundary conditions for the solvent flow imposed by surfaces of the colloidal particles and the container walls. Hence, $\mathbf{v}(\mathbf{r}')p(\mathbf{r}')$ depends implicitly on the position and orientations of all colloidal particles. Substitution of Eq. (24) into Eq. (23) leads to

$$\nabla\cdot\boldsymbol{\Sigma}^{ss}=\mathbf{M}^{(1)}+\mathbf{M}^{(2)}, \tag{25}$$

where

$$\mathbf{M}^{(1)}\equiv\lim_{\delta_x,\delta_y,\delta_z\to 0}\frac{\eta_0}{\delta_x\,\delta_y\,\delta_z}\times\left\langle\int_{A_s} d\mathbf{S}'\cdot[\nabla'\mathbf{v}(\mathbf{r}')+(\nabla'\mathbf{v}(\mathbf{r}'))^T]\right\rangle \tag{26}$$

and

$$\mathbf{M}^{(2)}\equiv-\lim_{\delta_x,\delta_y,\delta_z\to 0}\frac{1}{\delta_x\,\delta_y\,\delta_z}\left\langle\int_{A_s} d\mathbf{S}'\,p(\mathbf{r}')\right\rangle . \tag{27}$$

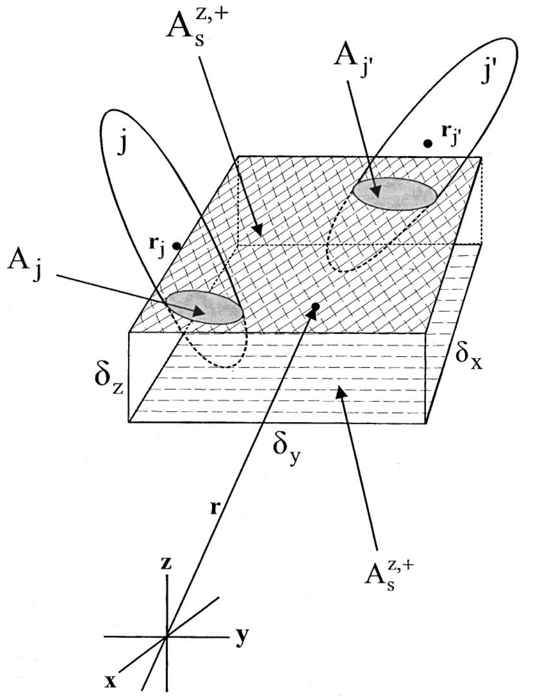

FIG. 2. The surface area $\partial\delta V$ of the volume element that is occupied by solvent is denoted as A_s. This is $\partial\delta V$ minus the areas A_j of intersection of $\partial\delta V$ with the core of colloidal particle j. The part of A_s on the upper z-side of the volume element is referred to as $A_s^{z,+}$, and on the lower z-side as $A_s^{z,-}$.

Consider first the contribution $\mathbf{M}^{(1)}$. We can rewrite the integral as

$$\left\langle\int_{A_s} d\mathbf{S}'\cdot[\nabla'\mathbf{v}(\mathbf{r}')+(\nabla'\mathbf{v}(\mathbf{r}'))^T]\right\rangle =\left\langle\left[\oint_{\partial\delta V}-\sum_{j=1}^{N}\int_{A_j}\right]d\mathbf{S}'\cdot[\nabla'\mathbf{v}(\mathbf{r}')+(\nabla'\mathbf{v}(\mathbf{r}'))^T]\right\rangle , \tag{28}$$

where A_j is the area of intersection of the surface area $\partial\delta V$ of the volume element and the core of colloidal particle j (see Fig. 2). For a rigid colloidal particle, the velocity inside the core is given by

$$\mathbf{v}(\mathbf{r}')=\mathbf{v}_j+\boldsymbol{\Omega}_j\times(\mathbf{r}'-\mathbf{r}_j),\qquad \mathbf{r}'\in\ \text{core of particle}\ \ j, \tag{29}$$

where $\mathbf{v}_j$ is the translational velocity and $\boldsymbol{\Omega}_j$ the rotational velocity of colloidal particle j. Hence (with $\boldsymbol{\epsilon}$ the Levi–Cevita tensor, and $\Omega_{j,p}$ the pth component of $\boldsymbol{\Omega}_j$),

$$\nabla'_m v_n=\nabla_m[\boldsymbol{\Omega}_j\times(\mathbf{r}'-\mathbf{r}_j)]_n=\nabla'_m\,\epsilon_{npq}\,\Omega_{j,p}\,r'_q =\epsilon_{npm}\,\Omega_{j,p},$$

where summation over repeated indices is assumed. From the antisymmetry of the Levi–Cevita tensor, it is thus found that

$$\int_{A_j} d\mathbf{S}'\cdot[\nabla'\mathbf{v}(\mathbf{r}')+(\nabla'\mathbf{v}(\mathbf{r}'))^T]=\mathbf{0}. \tag{30}$$

Using Gauss' integral theorem, we thus find from Eqs. (28) and (30), for incompressible solvents,

$$\left\langle \int_{A_s} d\mathbf{S}' \cdot [\nabla' \mathbf{v}(\mathbf{r}') + (\nabla' \mathbf{v}(\mathbf{r}'))^T] \right\rangle = \left\langle \int_{\delta V} d\mathbf{r}' \, \nabla'^2 \mathbf{v}(\mathbf{r}') \right\rangle .$$

Hence, Eq. (26) reduces to

$$\mathbf{M}^{(1)} = \lim_{\delta_x,\delta_y,\delta_z \to 0} \frac{\eta_0}{\delta_x \, \delta_y \, \delta_z} \left\langle \int_{\delta V} d\mathbf{r}' \, \nabla'^2 \mathbf{v}(\mathbf{r}') \right\rangle$$
$$= \eta_0 \langle \nabla^2 \mathbf{v}(\mathbf{r}) \rangle .$$

From Eq. (5), it is thus finally found that

$$\mathbf{M}^{(1)} = \eta_0 \, \nabla^2 \mathbf{U}(\mathbf{r},t). \tag{31}$$

The contribution $\mathbf{M}^{(2)}$ can be expressed in suspension properties as follows. Let $A_s^{z,+}$ denote the top-side of A_s, and similarly $A_s^{z,-}$ the lower-side, as indicated in Fig. 2. Furthermore, let $\hat{\mathbf{e}}_z$ denote the unit vector along the positive z-axis. Since the unit normal on $A_s^{z,+}$ is $\hat{\mathbf{e}}_z$ while the unit normal on $A_s^{z,-}$ is $-\hat{\mathbf{e}}_z$, the contribution $\mathbf{M}_z^{(2)}$ from the top- and lower-side of A_s to $\mathbf{M}^{(2)}$ is equal to

$$\mathbf{M}_z^{(2)} = -\hat{\mathbf{e}}_z \lim_{\delta_x,\delta_y,\delta_z \to 0} \frac{1}{\delta_x \, \delta_y \, \delta_z}$$
$$\times \left\langle \left[\int_{A_s^{z,+}} - \int_{A_s^{z,-}} \right] dS' \, p(\mathbf{r}') \right\rangle .$$

For small sizes of the volume element, the scalar,

$$P^{ss}(\mathbf{r} + \tfrac{1}{2} \delta_z \hat{\mathbf{e}}_z, t) \equiv \frac{1}{\delta_x \, \delta_y} \left\langle \int_{A_s^{z,+}} dS' \, p(\mathbf{r}') \right\rangle , \tag{32}$$

defines the contribution to the suspension pressure due solvent-solvent interactions at the position of the top-side of the volume element. A similar expression can be written down for P^{ss} at the lower-side. It thus follows that

$$\mathbf{M}_z^{(2)} = -\hat{\mathbf{e}}_z \lim_{\delta_z \to 0} \frac{1}{\delta_z} [P^{ss}(\mathbf{r} + \tfrac{1}{2} \delta_z \hat{\mathbf{e}}_z, t)$$
$$- P^{ss}(\mathbf{r} - \tfrac{1}{2} \delta_z \hat{\mathbf{e}}_z, t)]$$
$$= -\hat{\mathbf{e}}_z \frac{\partial P^{ss}(\mathbf{r},t)}{\partial z} .$$

In the same way the contribution to $\mathbf{M}^{(2)}$ from the left- and right-sides, and front- and back-sides of the volume element are obtained. Summing these contributions leads to

$$\mathbf{M}^{(2)} = -\nabla \, P^{ss}(\mathbf{r},t). \tag{33}$$

We thus find from Eqs. (25), (31), (33) the following expression for the divergence of the stress tensor that arises from solvent–solvent interactions:

$$\nabla \cdot \mathbf{\Sigma}^{ss} = \eta_0 \, \nabla^2 \mathbf{U}(\mathbf{r},t) - \nabla \, P^{ss}(\mathbf{r},t). \tag{34}$$

This expression is identical to that derived by Batchelor[8] for homogeneous systems.

Note that P^{ss} is not just determined by boundary conditions, contrary to p in Eq. (24), since the suspension is inhomogeneous, resulting in a spatially varying viscosity.

VI. THE DIVERGENCE OF THE TOTAL STRESS TENSOR

On the Smoluchowski time scale, the interaction force $\mathbf{F}_j$ in Eq. (9) balances with the hydrodynamic force in Eq. (20),[10,11] that is,

$$\mathbf{F}_j + \mathbf{F}_j^h = \mathbf{0}. \tag{35}$$

On using force balance, admissible values of the inverse local shear rate and time scales are bounded from below by the Smoluchowski time scale, which is of the order of $10^{-8} - 10^{-7}$ s. This is well within the range of shear rates of interest and time scales on which phase separation and shear-banding occurs in colloidal systems. The first term in Eq. (19) for the particle–solvent stress thus cancels against the particle–particle stress in Eq. (11). Adding Eqs. (11), (19), (34) thus leads to the following expression for the divergence of the total stress tensor:

$$\nabla \cdot \mathbf{\Sigma} = \eta_0 \, \nabla^2 \mathbf{U}(\mathbf{r},t) - \nabla P^{ss}(\mathbf{r},t)$$
$$- \sum_{j=1}^{N} \left\langle \oint_{\partial V_j(\mathbf{r}_j,\omega_j)} dS' \, \delta(\mathbf{r} - \mathbf{r}') \, \mathbf{f}^h(\mathbf{r}') \right\rangle . \tag{36}$$

This expression is the main result of the present paper. This seemingly simple expression is valid for suspensions with arbitrary large gradients in shear rate, concentration and orientational order parameters.

When evaluating this general expression explicitly in terms of shear rate, concentration and orientational order parameters, there are two possible ways to deal with the pressure. One can either separate all terms that can be written as $\sim \nabla f$, with f a scalar field, and identify $P^{ss} + f$ as the suspension pressure (which includes the osmotic pressure), or one can leave P^{ss} as it stands as an unknown scalar field. In both cases one has just as many equations as unknown fields.

The main result (36) has a quite straightforward interpretation. The last term on the right-hand side is simply the ensemble averaged force per unit volume that surface elements of colloidal particles exert on the fluid, provided that that surface element is located at $\mathbf{r}$ (as indicated by the delta distribution).

Like for homogeneous systems, divergent integrals are encountered when explicitly evaluating our formal result (36) for the stress tensor.[12] These divergencies are due to the long ranged nature of hydrodynamic interactions. Batchelor's arguments[12] to deal with these divergencies should be extended to inhomogeneous systems when Eq. (36) is evaluated for systems where hydrodynamic interactions are important, like hard sphere suspensions. This nontrivial problem is outside the scope of the present paper. In a forthcoming publication,[1] we shall evaluate Eq. (36) for suspensions of very long and thin, hard rods. Since volume fractions of interest scale like D/L (with D the thickness and L the length of the rods), hydrodynamic interactions are much less important for very long and thin rods as compared to hard sphere systems. Stresses are now predominantly generated by direct interactions rather than hydrodynamic interactions, so that divergence problems are not encountered.

Note that for the evaluation of Eq. (36), the solvent flow and pressure around each rigid, colloidal particle must be calculated in order to be able to calculate $\mathbf{f}^h(\mathbf{r}')$. This is precisely what is done in our forthcoming publication[1] for very long and thin rods.

VII. SUMMARY AND CONCLUSIONS

The main result of this paper is expression (36) for the divergence of the suspension stress tensor. This expression is a necessary ingredient to describe phenomena under shear flow where (possibly large) gradients in shear rate, concentration, and orientational order parameters are present.

In an application of Eqs. (1), (2), (36) to a particular system, the expression (36) should be expressed explicitly in terms of shear rate, concentration and relevant order parameters. In a future publication, this highly nontrivial step will be performed for suspensions containing very long and thin, uniaxial, rigid rods with hard-core interactions. In fact, it will be shown that the stress tensor (36) contains convolution-type integrals that involve the joint probability density function $\rho(\mathbf{r},\hat{\mathbf{u}},t)$ for the position coordinate $\mathbf{r}$ and the orientation $\hat{\mathbf{u}}$ of a rod. Hence, for systems of rods, an additional equation of motion for this joint probability density function must be derived to complete the set of Eqs. (1), (2), (36). This requires a separate analysis, independent from the one given in the present paper for the stress tensor. As for the stress tensor, the equation of motion for $\rho(\mathbf{r},\hat{\mathbf{u}},t)$ turns out to contain convolution-type integrals. Equations of motion that are of a similar structure as commonly used equations are recovered, when the convolution-type integrals are expanded in a spatial gradient series, and truncating after the fourth order derivatives. Such a truncation is questionable when gradients become large, in which case the convolution-type of integrals must be kept as they stand.

Since simple shear flow is a nonconservative external field, no Hamiltonian can be defined for systems in shear flow, and hence, no free energy can be defined. This obscures thermodynamic reasoning in deriving expressions for the stress tensor. The derivation of Eq. (36) for the stress tensor does *not* rely on thermodynamic reasoning, but is a mere summation of forces that act on a volume element.

The unknown scalar field P^{ss} (the contribution to the suspension pressure due to solvent–solvent interactions) can be dealt with in two ways. First of all, in an explicit evaluation of Eq. (36) for a particular system, one may identify all contributions of the form ∇f, with f a scalar field, and introduce the total suspension pressure P as $P=P^{ss}+f$. The second way to deal with P^{ss} is to keep it as it stands; just like the total pressure, P^{ss} is an *a priori* unknown field that can be found from the complete set of equations. For example, for the rigid rods discussed above, there are three equations for the three unknown fields $\mathbf{U}$, P^{ss}, and $\rho(\mathbf{r},\hat{\mathbf{u}},t)$; Eqs. (1), (2) [with an explicit expression for the stress tensor derived from Eq. (36)], and the equation of motion for $\rho(\mathbf{r},\hat{\mathbf{u}},t)$.

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APPENDIX A: ON THE VALIDITY OF EQ. (3)

Let $\mathbf{\Gamma}$ denote the phase space variables of the rigid colloidal particles. Let $\rho_m(\mathbf{r}|\mathbf{\Gamma})$ denote the microscopic mass density at position $\mathbf{r}$. This mass density is equal to that of the fluid when $\mathbf{r}$ is inside the solvent, and equal to that of the colloidal material when $\mathbf{r}$ is inside the core of a colloidal particle. Similarly, let $\mathbf{v}(\mathbf{r}|\mathbf{\Gamma})$ denote the microscopic velocity. This is the solvent flow velocity when $\mathbf{r}$ is inside the solvent and is the velocity of a colloidal material element otherwise. On averaging the microscopic Navier–Stokes equation in order to obtain the effective Navier–Stokes equation (1), the suspension flow velocity $\mathbf{U}(\mathbf{r},t)$ must be defined as[2–6]

$$\mathbf{U}(\mathbf{r},t)=\langle\rho_m(\mathbf{r}|\mathbf{\Gamma})\mathbf{v}(\mathbf{r}|\mathbf{\Gamma})\rangle/\langle\rho_m(\mathbf{r}|\mathbf{\Gamma})\rangle. \tag{A1}$$

The ensemble averaging brackets $\langle\cdots\rangle$ denote averages with respect to a probability density function $P(\mathbf{\Gamma},t)$ that complies with the particular ensemble under consideration. The ensemble average $\langle\rho_m(\mathbf{r}|\mathbf{\Gamma})\rangle$ is the macroscopic mass density ρ_m that appears in the Navier–Stokes equation (1).

Let $\chi_f(\mathbf{r}|\mathbf{\Gamma})$ denote the characteristic function for the solvent [$\chi_f(\mathbf{r}|\mathbf{\Gamma})=1$ when $\mathbf{r}$ is in the solvent and $=0$ otherwise]. Similarly, let $\chi_c(\mathbf{r}|\mathbf{\Gamma})$ denote the characteristic function for the colloidal material [$\chi_c(\mathbf{r}|\mathbf{\Gamma})=1$ when $\mathbf{r}$ is inside the core of a colloidal particle and $=0$ otherwise]. For any phase function $\mathbf{f}(\mathbf{r}|\mathbf{\Gamma})$ we can then write

$$\begin{aligned}\langle\rho_m(\mathbf{r}|\mathbf{\Gamma})f(\mathbf{r}|\mathbf{\Gamma})\rangle&=\int d\mathbf{\Gamma}P(\mathbf{\Gamma},t)\rho_m(\mathbf{r}|\mathbf{\Gamma})f(\mathbf{r}|\mathbf{\Gamma})\\&=\int d\mathbf{\Gamma}P(\mathbf{\Gamma},t)\\&\quad\times\{\rho_0\chi_f(\mathbf{r}|\mathbf{\Gamma})+\rho_c\chi_c(\mathbf{r}|\mathbf{\Gamma})\}f(\mathbf{\Gamma}),\end{aligned}$$

where ρ_0 is the solvent mass density and ρ_c that of the colloidal material. This can be rewritten as

$$\langle\rho_m(\mathbf{r}|\mathbf{\Gamma})f(\mathbf{r}|\mathbf{\Gamma})\rangle$$
$$=\rho_0\int d\mathbf{\Gamma}P(\mathbf{\Gamma},t)\left\{1+\frac{\rho_c-\rho_0}{\rho_0}\chi_c(\mathbf{r}|\mathbf{\Gamma})\right\}f(\mathbf{r}|\mathbf{\Gamma}),$$

where it is used that $\chi_f+\chi_c=1$. Since the local volume fraction $\varphi(\mathbf{r},t)$ of colloidal material is equal to

$$\varphi(\mathbf{r},t)=\int d\mathbf{\Gamma}P(\mathbf{\Gamma},t)\chi_c(\mathbf{r}|\mathbf{\Gamma}),$$

it follows that

$$\begin{aligned}\langle\rho_m(\mathbf{r}|\mathbf{\Gamma})f(\mathbf{r}|\mathbf{\Gamma})\rangle&\approx\rho_0\int d\mathbf{\Gamma}P(\mathbf{\Gamma},t)f(\mathbf{r}|\mathbf{\Gamma})\\&=\rho_0\langle f(\mathbf{r}|\mathbf{\Gamma})\rangle,\end{aligned}$$

whenever φ and/or $(\rho_c-\rho_0)/\rho_0$ are small. Under these conditions we thus have (with $\mathbf{f}=\mathbf{v}$),

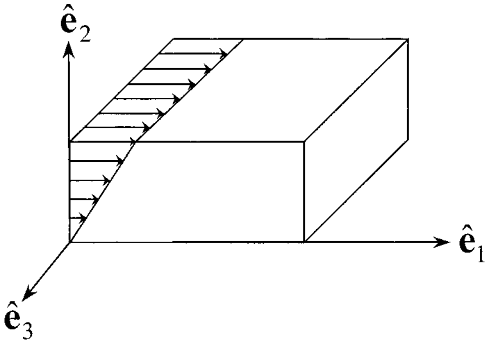

FIG. 3. A simple shear flow in the x-direction with its gradient in the y-direction.

$$\langle \rho_m(\mathbf{r}|\mathbf{\Gamma})\mathbf{v}(\mathbf{r}|\mathbf{\Gamma})\rangle \approx \rho_0 \langle \mathbf{v}(\mathbf{r}|\mathbf{\Gamma})\rangle,$$

and (with $f=1$)

$$\langle \rho_m(\mathbf{r}|\mathbf{\Gamma})\rangle \approx \rho_0,$$

so that Eq. (A1) for the suspension flow velocity reduces to Eq. (3).

For many real colloidal systems, the relative difference in mass density between solvent and colloidal material is indeed small, like for solvent-swollen microgel spheres and many biological macromolecules. In addition, for anisotropic colloidal particles, volume fractions of interest are usually small. For example, for long and thin rigid rods, the volume fractions of interest scale like D/L (with D the thickness of the rods and L their length), which is indeed a small number.

There is another good reason for approximating Eq. (A1) by Eq. (3), even when the two above conditions are not met. Contrary to more general two-phase flow, for colloidal systems the microscopic velocity varies relatively smoothly with position for any configuration of the colloidal particles. The reason for this is the overdamped dynamics of the colloidal particles; friction forces between colloidal particles and solvent dominate over inertial forces, so that colloidal particles attain much the same velocity as the surrounding solvent. Hence, for large mass density differences between solvent and colloidal particles, the mass density is a much more strongly varying function of $\mathbf{\Gamma}$ as compared to the velocity. This validates the factorization $\langle \rho_m \mathbf{v}\rangle \approx \langle \rho_m\rangle\langle \mathbf{v}\rangle$, which again reduces Eq. (A1) to Eq. (3).

APPENDIX B: Σ^{pp} FOR A HOMOGENEOUS SYSTEM

Consider a flow in the x-direction with its gradient in the y-direction, as depicted in Fig. 3. Since all suspension properties, including the suspension flow velocity, do not vary on xz-planes, the stress tensor is a function of y only. Hence,

$$\nabla \cdot \mathbf{\Sigma}(\mathbf{r}) = \hat{\mathbf{e}}_2 \cdot \frac{d\mathbf{\Sigma}(y)}{dy},$$

where $\hat{\mathbf{e}}_2$ is the unit vector in the y-direction. Since in a homogeneous system the force $\langle \delta(\mathbf{r}-\mathbf{r}_j)\,\mathbf{F}_j\rangle$ on rod j at $\mathbf{r}$ is independent of $\mathbf{r}$, it thus follows immediately from Eq. (11) by integration that

$$\hat{\mathbf{e}}_2 \cdot \mathbf{\Sigma}(y) = y \sum_{j=1}^{N} \langle \delta(\mathbf{r}-\mathbf{r}_j)\,\mathbf{F}_j\rangle = \hat{\mathbf{e}}_2 \cdot \sum_{j=1}^{N} \langle \delta(\mathbf{r}-\mathbf{r}_j)\,\mathbf{r}_j\,\mathbf{F}_j\rangle.$$

Here, the integration constant is set equal to 0, since it does not contribute to the force on a given volume element, and is therefore irrelevant. Since for the calculation of the shear viscosity only the component $\hat{\mathbf{e}}_2 \cdot \mathbf{\Sigma}$ of the stress tensor is of importance, it follows that expression (14) for the stress tensor can be utilized for such calculations.

APPENDIX C: Σ^{ps} FOR A HOMOGENEOUS SYSTEM

Since $\mathbf{r}' \in \partial V_j$ in the integrand in Eq. (19), the magnitude of $\mathbf{r}'-\mathbf{r}_j$ is never larger than the linear dimension of the rigid colloidal particles. Hence, for not too large gradients of suspension properties, the delta distribution $\delta(\mathbf{r}-\mathbf{r}')$ can be Taylor expanded around $\mathbf{r}'=\mathbf{r}_j$,

$$\delta(\mathbf{r}-\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}_j) + \sum_{n=1}^{\infty} \frac{1}{n!}(\mathbf{r}_j-\mathbf{r}')^n \odot \nabla^n \delta(\mathbf{r}-\mathbf{r}_j),$$

where $(\mathbf{r}_j-\mathbf{r}')^n$ and ∇^n are polyadic products, and $\odot$ is the n-fold contraction of these two products. On substitution of this expansion into Eq. (19), it is found that

$$\nabla \cdot \mathbf{\Sigma}^{ps} = -\sum_{n=1}^{\infty} \frac{1}{n!} \nabla^n \sum_{j=1}^{N} \left\langle \delta(\mathbf{r}-\mathbf{r}_j) \odot \oint_{\partial V_j(\mathbf{r}_j,\omega_j)} dS'\,(\mathbf{r}_j-\mathbf{r}')^n\,\mathbf{f}^h(\mathbf{r}') \right\rangle.$$

For homogeneous systems, only the leading order gradient term (the term for which $n=1$) survives, and hence,

$$\nabla \cdot \mathbf{\Sigma}^{ps} = \nabla \cdot \sum_{j=1}^{N} \left\langle \delta(\mathbf{r}-\mathbf{r}_j) \oint_{\partial V_j(\mathbf{r}_j,\omega_j)} dS'\,(\mathbf{r}'-\mathbf{r}_j)\,\mathbf{f}^h(\mathbf{r}') \right\rangle.$$

Since a divergenceless contribution to the stress tensor is of no significance, since the suspension flow velocity is determined solely through the divergence of the stress tensor, this result immediately leads to Eq. (21) for $\mathbf{\Sigma}^{ps}$ for a homogeneous system.

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