# HYDROMECHANICS OF BROWNIAN SUSPENSIONS

A. Yu. Zubarev and E. S. Kats

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The values of effective rheological characteristics and of the particle self-diffusion coefficient in concentrated Brownian suspensions are estimated.

The classical theory of Brownian particles [1] was developed for the limit of dilute suspensions, in which the interparticle interaction is assumed to be totally negligible. It is natural to expect that collective effects in concentrated systems, generated by Brownian motion near particle locations, can affect the rheological properties of the mixture and the characteristics of diffusion processes. This effect is analyzed below.

Rheological Properties. The effect of particle diffusion motion on rheological properties of suspensions was first investigated in [2], where it was shown that this motion enchances the system viscosity in comparison with the situation in which the particle chaotic displacements can be neglected. The analysis of [2], however, was carried out only for dilute mixtures, in which one can isolate two hydrodynamically interacting particles without taking into account the presence of remaining particles.

In most studies known to the present authors (see, for example, the review [3]) the problem of Brownian effects on the rheological properties of the suspension has generally not been treated. At the same time the results of [2] make it possible to expect that for concentrated systems these effects must be substantial. As a matter of fact, the experimentally determined dependences of the suspension viscosity on the disperse phase concentration, obtained by various authors, occupy a quite wide band in the $(\eta, \rho)$-plane (see Fig. 1). The most correct calculations of the function $\eta(\rho)$, carried out within models in which particle diffusion is neglected, are in the lower part of this band [4]. This spread of experimental data can be fully due to the apparent dependence of Brownian effects on experimental conditions. The effect of internal diffusion on the effective viscosity of moderately concentrated $(\rho \leqslant 0.2)$ suspensions was considered in [5]; here systems are investigated with an arbritrary disperse phase concentration.

The suspension is considered as a system of identical solid spheres, suspended in a Newtonian fluid. Many equivalent relations [2-4, 6] are known for determining $\eta$. We use here the representation following from the results of [6]:

$$4 (\eta - \eta_0)\, \mathbf{e} \cdot \mathbf{e} = \frac{3\rho}{4\pi a^3} \int\limits_{r=a} (\sigma_{rr}^+ u_r + \sigma_{r\theta}^+ u_\theta + \sigma_{r\varphi}^+ u_\varphi)\, d\mathbf{r}, \tag{1}$$

where $\mathbf{u}$ is the mean suspension velocity.

The $\mathbf{e}$ and $\mathbf{u}$ components are related as usual. In a Cartesian coordinate system, for example,

$$e_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial r_j} + \frac{\partial u_j}{\partial r_i} \right), \quad i,\, j = x,\, y,\, z. \tag{2}$$

The integration in (1) is carried out over a surface of an arbitrarily selected trial sphere. We emphasize that $\mathbf{e}$ and $\mathbf{u}$ correspond to suspensions in a homogeneous fluid, while $\sigma^+$ is determined with account of perturbations due to a test particle in the averaged flow.

The stress $\sigma^+$ is the sum of the hydrodynamic stress $\sigma_H^+$, formed by the perturbated flow near the test sphere, and the Brownian stress $\sigma_B^+$, accompanying the diffusion motion
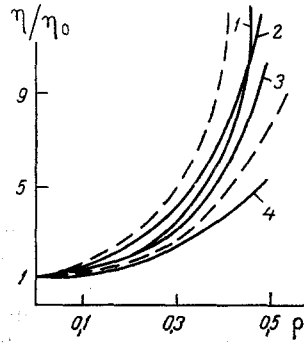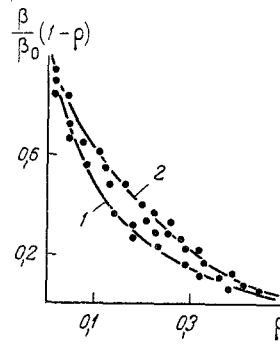
Fig. 1. Relative effective suspension viscosity as a function of the bulk concentration of the disperse phase: 1) the theory suggested; 2) the empirical equation of [9]; 3) calculations of [4] for the binary distribution function $n^0$ in the Percus—Yewick form; 4) calculation [4] for $\rho^0$ in the form (12); the dashed lines are the boundaries of the spread of experimental data, selected in [4].

Fig. 2. Relative mobility of spherical particles ($\beta_0 = (6\pi\eta_0 a)^{-1}$) as a function of their bulk concentration: 1) the theory suggested; 2) calculations of [4] for the binary distribution function $n^0$, selected in the Percus—Yewick form. The points are results of various experiments, included in [4].

of the disperse phase near the test particle [1, 5]. The physical reason for the generation of $\sigma_B^+$ is the fact that the convective flow destroys the statistical homogeneity of the particle distribution, while the diffusion motion tends to conserve it. Internal flows are generated in the suspension as a result, to overcome which it is necessary to apply supplementary forces.

The basic complexity of the physical mechanics of the mixture is determining the flows near the test particle, since in this case it is necessary to take into account the large number of remaining terms. Obviously, the most exhaustive analysis of this problem was carried out in [7], where it was shown that the hydrodynamic stress $\sigma_H^+$ can be determined from the solution of the problem:

$$-\nabla\sigma_H^+ = 0; \quad \operatorname{div}\mathbf{u}^+ = 0; \quad r > a; \quad \mathbf{u}^+ = 0, \quad r = a; \quad \mathbf{u}^+ \to \mathbf{u}, \quad r \to \infty;$$

$$\sigma_H^+ = -p_H^+ \mathbf{1} + 2\eta^+(\mathbf{e}^+ - \mathbf{e}) + 2\eta\mathbf{e}; \quad \eta^+(r) = \eta_0 + (\eta - \eta_0)\frac{\rho^+(r)}{\rho}, \tag{3}$$

where $\mathbf{e}^+$ and $\mathbf{u}^+$ are related as in (2). The function $\rho^+(r)$ is the conditional bulk concentration of the disperse phase near the test sphere. Its physical meaning is the probability of finding the point $r$ inside one of the particles if the test particle is fixed at the origin of coordinates. The shape of $\rho^+(r)$ for systems with a homogeneous distribution of the disperse phase was analyzed in [4], and is discussed briefly below.

In the theory of [7] the test sphere is considered as immersed in a fictitious continuous medium, whose properties are dictated by the shape of the conditional bulk concentration $\rho^+$. Repeating the considerations of [5], within this model we reach the following representation for the Brownian stress $\sigma_B^+$:

$$\sigma_B^+ = -p_B^+\mathbf{1}, \quad \nabla p_B^+ = n^+ \frac{\partial\mu(\rho^+)}{\partial\rho^+}\nabla\rho^+, \tag{4}$$

Taking into account that $D_e = \beta\partial\mu/\partial\rho$, and substituting $p_B^+$ from (10) and $\sigma_B^+$ into (1), we calculate the integral appearing there in the form of a nonlinear function of $\eta$ and $\beta$. Thus, Eq. (1) is transformed into an equation in $\eta$, containing $\beta$ as a parameter. To close the problem it is necessary to determine the particle mobility $\beta$. This quantity has been estimated numerous times by various authors [3] without taking into account diffusion motion. Starting from the definition of the hydrodynamic mobility, we have the following relation:

$$\beta^{-1}\mathbf{u} = \int\limits_{r=a} (\sigma_{rr}^+ \cos\theta - \sigma_{r\theta}^+ \sin\theta)\, dr, \qquad (11)$$

where the integration is carried out over the surface of the test sphere, and $\theta$ is the angle in a spherical coordinate system with the axis along $\mathbf{u}$.

The quantity $\sigma^+$ can now be determined on the basis of the discussion above, if one assigns the mean velocity $\mathbf{u}$ in (3) at an infinite distance from the test sphere. As a result we calculate the integral (11) in the form of a function of $\eta$ and $\beta$, obtaining the same equation in $\beta$, containing $\eta$ as a parameter. Thus, following these operations, Eqs. (1) and (11) are two nonlinear equations in $\eta$ and $\beta$, solving which we determine the rheological properties of the suspension within the approximation considered.

The realization of this program is rendered difficult by the complexity of solving (3) if $\rho^0(r)$ is selected in the form of a smooth function, such as (9). The calculations are simplified by neglecting particle nonoverlapping and putting $\rho^0 = \rho$. This approximation, assumed if $\rho$ is not too large (as a rule $\rho \leqslant 0.25$), was investigated in [5]. In analyzing higher concentration systems it is convenient to use step function approximations of $\rho(r)$. The simplest step function approximation $\rho^0$ is:

$$\rho^0 = \begin{cases} 0, & r \leqslant 2a, \\ \rho, & r > 2a. \end{cases} \qquad (12)$$

In this case (3) formally coincides with the problem of a particle placed in a continuous medium, equivalent in its properties to the suspension, and distinct from the test sphere by a concentric layer of thickness a, filled by a purely disperse fluid. To simplify the calculations we use the approximation $\rho' = \dfrac{4}{3}\pi a^3 n'$, which is similar to the representation (12) of the function (9). Taking into account (7), the solution (3), (6) is elementary [6], but the results have an awkward shape, and are therefore not provided.

Figures 1, 2 compare results of calculations by the theory suggested with results of various experiments and with the calculations of [4], carried out, as above, by the methods of [7], but without including Brownian stresses, corresponding to the case Br >> 1. We note that in [9] were given empirical equations for the dependence of $\eta$ on Br, but subsequent analysis of the function $\eta$ in a wide range of Br numbers, though known to the authors, is not provided.

There exist very few experimental studies of the dependence of rheological properties of dispersion on Br. The experimental data selected in the review [9] show a decrease in the dependence $\eta(Br)$, verifying the hypothesis of enhanced effective suspensions viscosity due to internal diffusion processes.

Self-Diffusion Coefficient. The thermal motion of Brownian particles is characterized by two diffusion coefficients. The first is the gradient diffusion coefficient $D_e$, relating the particle concentration gradient with the diffusion flow. The second is the self-diffusion coefficient $D_s$, determining the value of particle displacements during their random walk in an isotropic suspension. In the limit of dilute media $D_e$ and $D_s$ are equal [1], but this is not the case for concentrated systems. However, the nature of the $\rho$-dependence of $D_e$ and $D_s$ is different [8, 10].

The effect of collective effects on $D_e$ and $D_s$ was first investigated in [10], following which the $\rho$-dependence of $D_s$ was analyzed in [11-15]. Even though very dilute systems were considered in [10-15] ($\rho \ll 1$), and only binary particle interactions were included, the results of these studies are different, which is explained by the complexity of accounting

where $p_B^+$ means the pressure created at the surface and near the test sphere by particles performing convective–diffusive motion, and $\mu(x)$ is the chemical potential of the solid sphere system, whose bulk concentration is x. This quantity was estimated, for example, in [8]. The function $n^+$ is the numerical concentration of the disperse phase near the test particle, related to $\rho^+$ by the relation [7]:

$$\rho^+(\mathbf{r}) = \int\limits_{|\mathbf{r}-\mathbf{r}'|\leqslant 2a} n^+(\mathbf{r}')\,d\mathbf{r}'. \tag{5}$$

The concentration $n^+(r)$ can be determined by solving the problem of convective particle diffusion near the test sphere, being in the stationary case

$$\nabla D_e(n^+)\nabla n^+ - \mathbf{u}^+\nabla n^+ = 0, \quad r > 2a;$$

$$D_e(n^+)\frac{\partial}{\partial r}n^+ + n^+u_r^+ = 0, \quad r = 2a; \quad n^+ \to n, \quad r \to \infty. \tag{6}$$

The first boundary condition of (6) follows from particle nonoverlapping, due to which the radial component of the disperse phase flow vanishes at the test sphere for r = 2a. The second is a consequence of the principle of correlation attenuation. The diffusion coefficient $D_e$ was estimated in [8].

The problem (3)-(6) is substantially nonlinear. Its solution is simplified for small Brenner Br = $ea^2/D_e$ or Peclet Pe = $ua/D_e$ numbers, when diffusive motion plays a decisive role in the formation of the disperse phase distribution. Precisely these situations are considered below.

We put

$$n^+ = n^\circ + n'; \quad \rho^+ = \rho^\circ + \rho'; \quad \frac{\rho'}{\rho^\circ}, \frac{n'}{n^\circ} \sim \text{Br}, \text{ Pe} \ll 1. \tag{7}$$

The quantity $n^0$ can be determined by solving independent problems of statistical physics. The simplest variant of $n^0$, taking into account particle nonoverlapping, is

$$n^\circ = \begin{cases} 0, r \leqslant 2a, \\ n \equiv \dfrac{3\rho}{4\pi a^3}, \ r > 2a. \end{cases} \tag{8}$$

The corresponding value of the equilibrium condition of the bulk concentration $\rho^0$, determined from (5), is such that:

$$\rho^\circ = 0, \ \xi < 1; \ \rho^\circ = \frac{27 - 56\xi + 30\xi^2 - \xi^4}{16\xi}\rho, \ 1 < \xi < 3,$$

$$\rho^\circ = \rho, \ \xi > 3; \ \xi = \frac{r}{a}. \tag{9}$$

Other variants of $n^0$ and $\rho^0$ were considered in [4]. It can be shown that only those parts of the hydrodynamic stress $\sigma_H^+$, determined from (3) for $\rho^+ = \rho^0$ without account of $\rho'$, provide a contribution to the integral (1).

The calculate $\eta$ we assign the mean tensor of suspension flow velocity e. Putting $\mathbf{u} = \mathbf{e}\cdot\mathbf{r}$, $\rho^+ = \rho^\circ$, from (3) we calculate $\sigma_H^+$ and $\mathbf{u}^+$. Further, from the linearized variant of (6) we determine $n'$. Calculating $\rho'$ rrom (5), within the linear approximation in Br we find from (4)

$$p_B^+ = n^\circ \frac{\partial\mu(\rho^\circ)}{\partial\rho^\circ}\rho'. \tag{10}$$
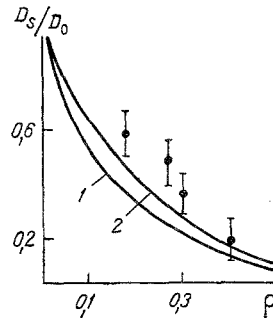
Fig. 3. Comparison of calculation results of the self-diffusion coefficient of spherical Brownian particles by the theory suggested with experimental data [15]: 1) $\beta$ selected in the form suggested by the present theory; 2) in the form [4] for $n^0$ selected in the Percus–Yewick form. The parameter $D_0$ is the diffusion coefficient of a single particle.

even for the binary interaction of chaotically moving spheres. A concentrated suspension of Brownian particles can be considered as a dense gas with a hydrodynamic noncentral and non-conservative interaction. A rigorous investigation of such a system is encountered with insurmountable mathematical difficulties. Therefore, at the present phase of analyzing Brownian motion in concentrated systems it is advisable to use approximations having a clear physical meaning, which may need to be made more complex while allowing substantial simplification of the computation.

The Langevin equation describing the motion of an arbitrary test Brownian test particle can be written in the form:

$$m\frac{d\mathbf{u}}{dt} = \mathfrak{f}_H + \mathfrak{f}_T, \tag{13}$$

where m and u are its mass and velocity.

The hydrodynamic force $\mathfrak{f}_H$ depends on the particle motion velocity in the field of random flows, created by other moving particles. These flows are generated, firstly, during reflection of perturbations accompanying a test sphere moving away from neighboring particles, and, secondly, directly by chaotically displaced particles surrounding the one considered. The first effects are similar to the collective effects generated in a regularly moving suspensions, and reduce the disperse phase mobility. The second ones lead to further chaotic particle motion. It can be expected that this generates an increase in the self-diffusion coefficient.

In analyzing the motion of a Brownian particle it is necessary to take into account that the random walk of neighbors screens it from the effect of the remaining particles. In this case the problem of determining $\mathfrak{f}_H$ can be solved by means of the known reflection method [3, 10]; this path, however, leads to very awkward final relations. To simplify the calculations we restrict ourselves to the first step of this method only, considering the test sphere in the hydrodynamic field of nearest neighbors, whose motion is not perturbed by the effect of the test and of the remaining particles. For regularly moving dense suspensions this approximation is too crude, but the statistical nature of the behavior of Brownian particles renders it more valid. This is stated in more detail below.

If, as assumed below, the conditions are realized for which one can neglect the inertial terms in the expression for $\mathfrak{f}_H$, then within the approximations made

$$\mathfrak{f}_H = \beta^{-1}(\mathbf{U} - \mathbf{u}), \tag{14}$$

where    is the total fluid velocity, created at the point of finding the nearest neighbors of the test particle.

Taking into account that the viscous relaxation time of Brownian particle motion is shorter than the propagation time of the hydrodynamic perturbation at scales of order a, nearest particles to the test particle are assumed to be uniformly moving. In this case

$$\mathbf{U} = \sum_j \int_0^\infty \mathbf{V}_j(\mathbf{r},\ t)\,\varphi_{ij}(\mathbf{r})\,N(\mathbf{r})\,d\mathbf{r},\ i,\ j = x,\ y,\ z. \tag{15}$$

The function $N(\mathbf{r})$ can be assigned by the well known nearest neighbor distribution [17], but the calculations become very awkward in this case. To obtain estimation results allowing to analyze the essence we assyne that the particles nearest to the test particle are distributed with density $n^0$ inside a coordination sphere of radius $2a\rho^{-1/3}$. This approximation is similar to that often used in solid state theory [18]. The approximation (8), (15) selected for $n^0$ is rewritten as:

$$U_i = \sum_j n \int_{2a}^{2a\rho^{-1/3}} V_j(\mathbf{r},\ t)\,\varphi_{ij}(\mathbf{r})\,d\mathbf{r}. \tag{16}$$

The function $\varphi_{ij}$ is derived in almost all textbooks of hydrodynamics (see, for example, [16]). Equation (14) is further analyzed by methods of the theory of correlation functions [19], according to which each random quantity h can be represented in the form of a Fourier–Stieltjes integral with a random Wiener measured $Z_h$:

$$h(\mathbf{r},\ t) = \int \exp(i\omega t + i\mathbf{kr})\,dZ_h,\ i = \sqrt{-1}, \tag{17}$$

while the correlator of the quantitites h and g can be expressed as:

$$\langle\, h(t+\tau,\ \mathbf{r}+\mathbf{x}),\ g(t,\ \mathbf{r})\,\rangle = \int \exp(i\omega t + i\mathbf{kr})\,\Psi_{hg}(\omega,\ \mathbf{k})\,d\omega d\mathbf{k},$$

$$\Psi_{hg} = \frac{\langle\, dZ_h dZ_g^*\,\rangle}{d\omega d\mathbf{k}}, \tag{18}$$

where the asterisk denotes complex conjugation.

Representing the random functions (14) in the form of Fourier–Stieltjes integrals, we obtain, for example:

$$mi\omega dZ_{u_z} = \beta^{-1}(\rho\gamma - 1)\,dZ_{u_z} + dZ_T,\ \rho\gamma = n\int_{2a}^{2a\rho^{-1/3}}\varphi_{zz}(\mathbf{r})\exp(i\mathbf{kr})\,d\mathbf{r},\ dZ_T \equiv dZ_{fT}, \tag{19}$$

if the direction of k is selected to be the z axis. In deriving (19) it was taken into account that all particles are equivalent, and therefore $dZ_V = dZ_u$. We note that it is precisely the statistical equivalence of particles which allows not to include in (15) the effect of all particles on the motion of nearest neighbors toward the test sphere, since this effect is self-consistently accounted for by the equality of measures $dZ_V$ and $dZ_u$.

Using for $\varphi_{zz}$ the known results of [16], we obtain

$$\rho\gamma = -\frac{3}{2a^3}\,n\int_{2a}^{2a\rho^{-1/3}} r^2 dr \int_{-\pi}^{\pi} \sin\theta d\theta \left\{\exp(ikr\cos\theta) \times\right.$$

$$\left. \times \left[1 - \frac{3}{4}(1+\cos^2\theta)\frac{a}{r} + \frac{a^3}{4r^3}(3\cos^2\theta - 1)\right]\right\}. \tag{20}$$

We further have from (19)

$$dZ_{u_z} = \frac{dZ_T}{\beta^{-1}(1-\gamma\rho) + mi\omega}. \tag{21}$$

By definition the self-diffusion coefficient $D_s$ equals:

$$D_s = \frac{1}{2}\lim_{t\to\infty}\frac{\langle z(t)z(0)\rangle}{t},$$

where z is the Cartesian coordinate of the mixture. Due to constraining effects the measure of $\|z\|$ of the set $\{z\}$ does not coincide with the measure $\|Z\|$ of the set $\{Z\}$ of all points of the medium. Introducing, as is usually done in the theory dense gases, the assumed volume $w \approx (1 - \rho/\rho_*)W$, where W is the volume of the whole mixture, and $\rho_* \approx 0.7$ is the dense concentration, we arrive at the estimate $\|z\| \approx (1-\rho/\rho_*)^{1/3}\|Z\|$. Using further the known procedure of deriving the Green-Kubo equation [20], we obtain

$$D_s \approx \pi \left(1 - \frac{\rho}{\rho_*}\right)^{-\frac{2}{3}}\int \Psi_{u_z u_z}(0,\ k)\,dk. \tag{22}$$

From (18) and (21) we have

$$\Psi_{u_z u_z} = \frac{\Psi_{TT}}{\beta^{-2}(1-\gamma\rho)^2 + m^2\omega^2}. \tag{23}$$

To determine $\Psi_{TT}$ we take into account that $f_T$ possesses properties of white noise [20], while the action of fluid molecules on a particle at different positions is uncorrelated. Therefore $\langle f(t,\ r)f(0,\ 0)\rangle = \alpha\delta(t)\delta(r)$ where $\delta$ is a delta-function. Using in (18) the vanishing of this correlator, we obtain

$$\Psi_{TT} = \frac{\alpha}{2\pi}, \tag{24}$$

where $\alpha$ is a so-far undetermined parameter. To calculate it we use the theorem of equally distributed energy over degrees of freedom, whence

$$\langle u_z^2\rangle = \frac{3}{2}\frac{T}{m}. \tag{25}$$

Hence and from (23) it follows that

$$\frac{T}{m} = \int \frac{\Psi_{TT}(\omega,\ k)}{\beta^{-2}(1-\gamma\rho)^2 + m^2\omega^2}\,d\omega dk. \tag{26}$$

Substituting (24) into (26), we obtain an expression for $\alpha$:

$$\alpha = \frac{\beta^{-1}}{2\pi}JT, \quad J = \left(\int_0^\infty \frac{k^2 dk}{1-\rho\gamma}\right)^{-1}. \tag{27}$$

Using now relations (23), (24), and (27) in (22), we reach the result:

$$D_s = \beta T\left(1 - \frac{\rho}{\rho_*}\right)^{2/3}FJ, \quad F = \int_0^\infty \frac{k^2 dk}{(1-\rho\gamma)^2}. \tag{28}$$

Calculations show that FJ = 1. The factor FJ in (28) describes the stimulating effect of chaotic motion of neighbors on Brownian particle displacements. The equality FJ = 1 implies that there exists no such stimulation, while the effect of collective effects reduces to lower particle mobility and generation of exclusive volume effects. Analysis shows that this conclusion does not change for any choice of N in (15), as well as by including inertial

terms in (14). We note that in estimation $f_H$ in (14) we included above the basic components of this force. Therefore, the random flows created by neighboring particles cannot strongly affect the coefficient $D_S$; the problem of a more refined effect remains, however, open.

Figure 3 shows a comparison of $D_S$ calculations by (28) with experiment [15]. Taking into account the large number of assumptions made, the agreement between theory and experiment can be considered to be quite good.

## NOTATION

Here a denotes the particle radius, $e$ is the mean value of the velocity tensor of suspension shear flow, $f_H$ and $f_T$ are the hydrodynamic and thermal interaction forces of a Brownian particle with the fluid, $I$ is a second unit tensor, $k$ is the wave vector, n is the mean number concentration of the disperse phase, N is the distribution density of nearest neighbors, p is the pressure, r is the radius-vector, directed from the center of the test sphere, t is time, $V(r, t)$ is a random particle velocity, next to the test particle, found at moment t at the point $r$, $\beta$ is the hydrodynamic particle mobility with account of their interaction, $\eta_0$ is the viscosity of the dispersion fluid, $\eta$ is the effective suspension viscosity, $\rho$ is the bulk concentration of the disperse phase, $\sigma$ is the stress, $\varphi_{ij}(r)$ is a function relating the i-th component of $V(r, t)$ with the j-th component of the flow velocity, accompanying this neighboring particle at the origin of coordinates, and $\Psi_{hg}$ denotes the spectral density of processes h and 8. The subscripts r, $\theta$, $\varphi$ ,..., denote components of vectors and tensors in a spherical coordinate system with origin at the center of the test sphere, the superscript _ is the mean value of a quantity near the test particle, $^0$ and $'$ are the equilibrium and perturbed values of convective particle flow, and ($\cdot$) denotes dot product.

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