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On the influence of the particles–fluid interaction on the turbulent diffusion in a suspension

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Abstract

A theory for the turbulent diffusion of particles suspended in an incompressible fluid is presented. Special attention is paid to the influence on the turbulent diffusion of the two-way coupling between the carrier fluid and the dispersed particles. Also the influence of the hydrodynamic interaction between the particles and the influence of the finite-particle size are investigated. It is shown that the influence of the particles–fluid interaction and of the finite-particle size are significant. The hydrodynamic interaction between the particles appears to be negligible up to a volume fraction of 0.1.

Keywords: Particles; Two-phase flow; Particle–turbulence interaction; Turbulent diffusion

1. Introduction

An important issue in the development of calculation methods for turbulent fluid–particles flows is the influence of the particles on the turbulence of the carrier phase. In the review paper by Crowe et al. (1996) much information is given about the literature on this two-way coupling effect. Squires and Eaton (1990) investigated the modification of turbulence by small particles using direct numerical simulations of stationary, isotropic turbulence. Elgobashi and Truesdell (1993), using direct numerical simulations, studied the modification of decaying, homogeneous turbulence due to its interaction with dispersed small solid particles. Portela et al. (1998) used large-eddy simulation to investigate a channel flow laden with small heavy particles. Elgobashi and Abou-Arab (1983) developed a two-equation turbulence model for predicting two-phase flows (describing fluid and particles as a continuum). Lun and Liu (1997) applied a two-equation

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turbulence model too. They simulated the solid phase by using a Lagrangian approach, in which the particles trajectories and velocities are determined by integrating the particle equation of motion. Very recently Crowe (2000) introduced a new model for the carrier-phase turbulence based on the volume-averaged equation for the kinetic energy of the carrier phase.

Also several physical models have been proposed for the turbulence modification by particles. For instance Yarin and Hetsroni (1994) proposed a model for the particles–turbulence interaction, in which both turbulence suppression by the fine particles and turbulence enhancement by the coarse particles are considered. Yuan and Michaelides (1992) presented also a model for the turbulence modification in particle-laden flows based on the interaction between particles and turbulence eddies. Kenning and Crowe (1997) developed a model for carrier-phase turbulence in gas–particles flows. The model suggests the importance of interparticle spacing in establishing a turbulence length scale in particles–gas suspensions.

A different type of physical model to investigate the turbulence modification by particles has been developed by Felderhof and Ooms (1989, 1990), further extended by Felderhof and Jansen (1991) and Ooms and Jansen (2000). They developed a model for the dynamics of a dispersion of solid spherical particles in an incompressible viscous fluid, in particular, paying attention to the influence of the particles–fluid interaction on the effective transport coefficients of the fluid–particles suspension. A key point in their work is, that the flow considered is an unsteady Stokes flow. The undisturbed flow, without particles, is assumed to satisfy the equation

$$\rho_{\mathrm{f}} \frac{\partial \mathbf{v}_0}{\partial t} = -\nabla p + \eta \nabla^2 \mathbf{v}_0 + \mathbf{F}_0,$$

in which $\mathbf{v}_0$ is the undisturbed fluid velocity, p the fluid pressure, ρ_{f} the fluid density and η the fluid viscosity. $\mathbf{F}_0$ is the random forcing term, used to generate a pseudo-turbulent flow with a given energy spectrum. The linearity of the equation allows to incorporate earlier results by Felderhof and Ooms (1989) and Felderhof and Jansen (1991) on low Reynolds number suspensions with the pseudo-turbulent flow, and to include multi-particle interactions. A summary of the model is given in Appendix A. The linearity of the model is implicit in the formulation given in the appendix. The original papers should be consulted for further details.

In this paper, we use this physical model to study the diffusion of particles in a turbulently flowing suspension. A theory of turbulent diffusion is formulated in which account is taken of the effect of the particles on the flow and of the hydrodynamic interactions between the particles. Also the influence of the finite-particle size will be incorporated in the theory; in this respect it is an extension of the earlier work by Felderhof and Ooms (1990), in which the particles are treated as point particles. As in this earlier work we use a continuum picture and consider the time evolution of the particle number density in an Eulerian description. In this picture the particle number density satisfies a stochastic differential equation from which we derive a generalized diffusion equation.

In order to be able to solve the generalized diffusion equation the local mean particle velocity field has to be known. Due to friction and inertia the local mean particle velocity field differs from the local mean fluid velocity field. For that reason we use the above-mentioned physical model (see Appendix A) to determine the ratio of the local mean particle velocity field and the local mean fluid velocity field as function of the relevant dimensionless groups. These groups are the volume fraction of the particles, the ratio of the fluid density and the particle density, and the ratio of the particle diameter and the turbulence integral length scale. Starting from a postulated fluid turbulence spectrum in the absence of particles, the physical model predicts the corresponding fluid turbulence spectrum in the presence of particles and the particles turbulence spectrum. There are two versions of this physical model. The first one uses a point-force approximation for the particles and takes into account the hydrodynamic interaction between the particles. The second version accounts for the detailed flow around the finite-diameter particles, but does not consider their hydrodynamic interaction. We use the calculated turbulence spectrum of the particle velocity field for the solution of the generalized diffusion equation. In particular, the time-dependent behaviour of the turbulent diffusion coefficient of the particles and of the mean square displacement (of a cloud of marked particles in the suspension) will be determined as function of the same dimensionless groups mentioned earlier. Both the point-particle version and the finite-diameter particle version of the physical model are applied.

The diffusion of small particles in homogeneous, isotropic turbulence in the limit of an infinitely small particle volume fraction has been studied for some time. In this limit there is no influence of the particles on the turbulent fluid flow field. So the two-way coupling between the carrier fluid and the particles is absent. For such very dilute systems the early result of Tchen (1947) was that the long-term particle diffusivity would be uneffected by particle inertia and be the same as the diffusivity of Lagrangian fluid elements. However, Pismen and Nir (1978) and Reeks (1977, 1991) showed that there is a net effect of particle inertia on the long-term diffusivity. As mentioned the emphasis of the present paper is to study the influence of the two-way coupling effect on the turbulent diffusivity of the particles. For that reason no attention is paid to the limiting case of infinitely small particle volume fraction. However, the theoretical model used in this paper can also be applied to this limiting case. In the near future we hope to report on a comparison between predictions made with our model and the results of Pismen and Nir (1978) and of Reeks (1977, 1991).

2. Turbulent diffusion in a suspension

We consider a turbulently flowing fluid suspension consisting of an incompressible fluid in which a large number of particles are immersed. On the continuum level the distribution of particles is described by the number density $n(\mathbf{r},t)$, in which $\mathbf{r}$ is the place coordinate and t is the time. The rate of change of the number density is given by the balance equation

$$\frac{\partial n}{\partial t} + \nabla \cdot \mathbf{j} = s, \tag{1}$$

where $\mathbf{j}(\mathbf{r},t)$ is the particle current density, and $s(\mathbf{r},t)$ is the source density describing possible creation and absorption of particles. We assume that the current density may be written as a sum of two terms

$$\mathbf{j} = -D_{\mathrm{m}}\nabla n + n\mathbf{u}, \tag{2}$$

where D_{m} is the molecular particle diffusivity, and $\mathbf{u}$ is the local mean particle velocity. We assume that in the suspension at rest the particle density is uniform and given by a constant n_0. Thus, in a flowing suspension the particle density may be written as

$$n(\mathbf{r},t) = n_0 + n_1(\mathbf{r},t), \tag{3}$$

where the deviation from uniformity $n_1(\mathbf{r},t)$ may be due to a nonvanishing source density $s(\mathbf{r},t)$, or to an initial density disturbance $n_1(\mathbf{r},0)$. From (1)–(3) we find

$$\frac{\partial n_1}{\partial t} - D_{\mathrm{m}}\nabla^2 n_1 + \nabla\cdot(n_1\mathbf{u}) = s, \tag{4}$$

where we have used $\nabla\cdot\mathbf{u} = 0$, which can be proven to be consistent with our assumption concerning the incompressibility of the fluid and the nature of the coupling between the two constituents. (The statement $\nabla\cdot\mathbf{u} = 0$ is false for general particle systems at finite Reynolds numbers. It is well known that heavy particles tend to cluster in regions of high strain rate in a turbulent flow. But the statement is appropriate for the model system considered here.) The balance equation (4) is a stochastic differential equation of the type

$$\frac{\partial n_1}{\partial t} - \mathbf{A}_0 n_1 - \mathbf{A}_1 n_1 = s, \tag{5}$$

where $\mathbf{A}_0$ is a known operator and $\mathbf{A}_1$ depends on a stochastic ensemble. In this case $\mathbf{A}_0 = D_{\mathrm{m}}\nabla^2$, and $\mathbf{A}_1$ is an integral operator with kernel given by

$$\mathbf{A}_1(\mathbf{r},\mathbf{r}',t) = -\nabla\cdot[\mathbf{u}(\mathbf{r},t)\delta(\mathbf{r}-\mathbf{r}')] \tag{6}$$

with the stochastic velocity field $\mathbf{u}(\mathbf{r},t)$. We consider a state of homogeneous, (stochastic) stationary and isotropic turbulence for the suspension in a coordinate system in which the mean flow vanishes. It can be shown (see Appendix B) that to second order in $\mathbf{A}_1$ the equation for the average density is given by

$$\left[\frac{\partial}{\partial t} - \mathbf{A}_0 - \langle\mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\right]\langle n_1\rangle = s, \tag{7}$$

where the Green function $\mathbf{G}_0$ is given by $\mathbf{G}_0 = [\frac{\partial}{\partial t} - \mathbf{A}_0]^{-1}$. The angle brackets indicate an average over the stochastic ensemble. When we consider the limit of vanishing D_{m}, Eq. (7) becomes after Fourier transformation with respect to time (see Appendix C):

$$-\mathrm{i}\omega\langle n_{1\omega}(\mathbf{r})\rangle - \mathbf{D}_{\mathbf{u}}(\omega)\nabla^{\mathbf{2}}\langle\mathbf{n}_{\mathbf{1}\omega}(\mathbf{r})\rangle = \mathbf{s}_\omega(\mathbf{r}) \tag{8}$$

with the frequency-dependent diffusion coefficient for the particles given by

$$D_u(\omega) = \int_0^\infty \mathrm{e}^{\mathrm{i}\omega t}\mathbf{\Gamma}_u(0,t)\,\mathrm{d}t. \tag{9}$$

$\mathbf{\Gamma}_u(0,t)$ is the local time-dependent correlation function for the particle velocity field. ω is the frequency. After Fourier transformation with respect to place the diffusion coefficient can be written as (see Appendix D)

$$Re\,D_u(\omega) = \frac{8\pi^2}{3}\int_0^\infty q^2 S_u(q,\omega)\,\mathrm{d}q, \tag{10}$$

where $S_u(q,\omega)$ is the spectral density of the turbulent fluctuations of the particle velocity field $\mathbf{u}$; q is the wave number. As explained in Section 1 this particle turbulence spectrum can be calculated with the aid of the physical model (see Appendix A) for the dynamics of a suspension. Eq. (9) yields

$$Re\,D_u(\omega) = \int_0^\infty \boldsymbol{\Gamma}_u(0,t)\cos\omega t\,\mathrm{d}t, \tag{11}$$

this may be reverted to yield

$$\boldsymbol{\Gamma}_u(0,t) = \frac{2}{\pi}\int_0^\infty Re\,D_u(\omega)\cos\omega t\,\mathrm{d}\omega. \tag{12}$$

The longitudinal correlation is defined as

$$f_u(0,t) = \frac{\boldsymbol{\Gamma}_u(0,t)}{\frac{1}{3}\langle\mathbf{u}^2\rangle}. \tag{13}$$

3. Effective diffusion coefficient

Eq. (5) may also be solved for $s = 0$ as an initial value problem for given $n_1(\mathbf{r},0)$. We consider in particular

$$n_1(\mathbf{r},0) = \delta(\mathbf{r}). \tag{14}$$

We define the mean square displacement

$$\Delta(t) = \int \mathbf{r}^2\langle n_1(\mathbf{r},t\,|\,\mathbf{0},0\rangle\,\mathrm{d}\mathbf{r}, \tag{15}$$

where $\langle n_1(\mathbf{r},t\,|\,\mathbf{0},0\rangle$ is the average density under the condition $n_1(\mathbf{r},0) = \delta(\mathbf{r})$. One can then prove (see Appendix E) that

$$\Delta(t) = 6\int_0^t D_{u,\mathrm{eff}}(t')\,\mathrm{d}t' \tag{16}$$

with

$$D_{u,\mathrm{eff}}(t) = \int_0^t \boldsymbol{\Gamma}_u(0,\tau)\,\mathrm{d}\tau. \tag{17}$$

As explained in the previous section $\boldsymbol{\Gamma}_u(0,t)$ can be calculated, and therefore $D_{u,\mathrm{eff}}(t)$ can be determined from Eq. (17) and the mean square displacement $\Delta(t)$ from Eq. (16).

For the so-called long-term effective turbulent diffusion coefficient the following relation holds:

$$D_{u,\mathrm{eff}}(\infty) = D_u(\omega = 0). \tag{18}$$

Using Eq. (18) two additional remarks can be made. First, since in the point-particle approximation for $\omega = 0$ the velocity fields $\mathbf{u}$ (for the particle velocity field), $\mathbf{v}$ (for the fluid velocity field with particles) and $\mathbf{v}_0$ (for the fluid velocity field without particles) become identical, we expect the long-term effective diffusion coefficients $D_{u,\mathrm{eff}}(\infty)$, $D_{v,\mathrm{eff}}(\infty)$ and $D_{v_0,\mathrm{eff}}(\infty)$ also to be identical. In the finite-particle approach this will not be the case, since here the different velocity fields will have different values for $\omega = 0$. Second, since the long-term effective diffusion coefficients are coupled to the spectra at $\omega = 0$, for which inertial effects are absent, the coefficients $D_{u,\mathrm{eff}}(\infty)$, $D_{v,\mathrm{eff}}(\infty)$ and $D_{v_0,\mathrm{eff}}(\infty)$ are expected to be independent of the ratio of fluid density and particle density. In the following we will use these facts to check our theoretical predictions.

4. Results

To get a quantitative insight concerning the influence that turbulence modification by particles has on the turbulent diffusion in a suspension, a number of calculations was carried out. In this section, their results will be presented. The calculation procedure was as follows:

1. choose values for the dimensionless groups (the volume fraction of the particles, the ratio of the fluid density and the particle density, and the ratio of the particle diameter and the turbulence integral length scale);
2. choose a turbulence spectrum for the fluid velocity field without particles;
3. calculate the turbulence spectrum for the particle velocity field and for the fluid velocity field with particles using the physical model for the dynamics of the suspension in point-particle approximation and in finite-diameter-particle approximation (see Appendix A);
4. calculate $Re D_u(\omega)$ using Eq. (10), calculate $\mathbf{\Gamma}_u(0,t)$ using Eq. (12), calculate $D_{u,\text{eff}}(t)$ using Eq. (17), and finally calculate $\Delta(t)$ with Eq. (16).

In this paper, we concentrate on the turbulent diffusion; so we only give results for $D_{u,\text{eff}}(t)$ as function of the relevant dimensionless groups and we present results for $\Delta(t)$. No results will be presented for $Re D_u(\omega)$ or for $\mathbf{\Gamma}_u(0,t)$. We have made $D_{u,\text{eff}}(t)$ dimensionless by dividing it by $D_{v_0,\text{eff}}(\infty)$. $\Delta(t)$ is written in the following dimensionless form: $\Delta(t)v/\Lambda^2 D_{v_0,\text{eff}}(\infty)$, in which v is the kinematic viscosity of the fluid.

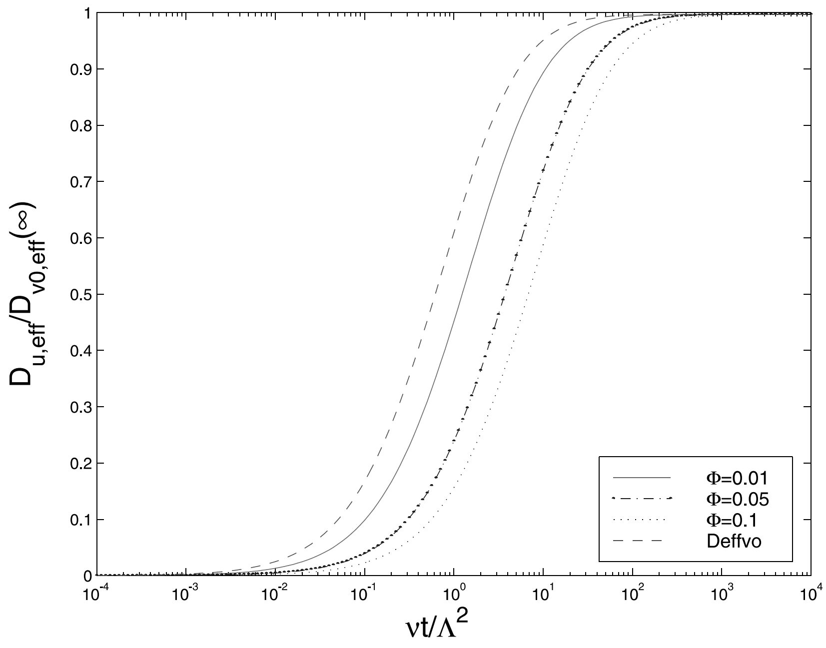

Fig. 1. Effective dimensionless diffusion coefficient for the particles as function of dimensionless time for several values of the particle volume fraction ϕ, and for $\rho_f/\rho_p = 0.01$ and $a/\Lambda = 0.01$ (point-particle approach). As a reference also $D_{v_0,\text{eff}}/D_{v_0,\text{eff}}(\infty)$ is shown.

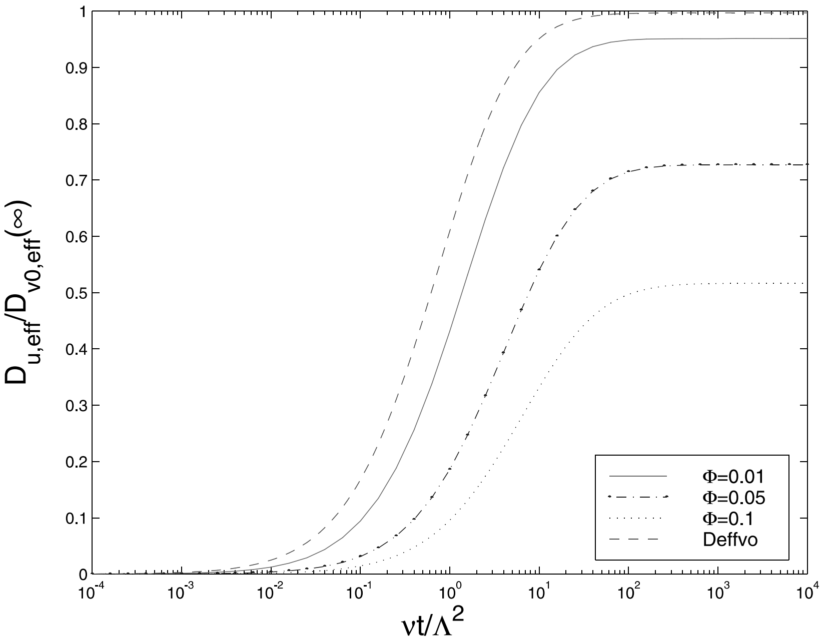

Fig. 2. Effective dimensionless diffusion coefficient for the particles as function of dimensionless time for several values of the particle volume fraction ϕ, and for $\rho_f/\rho_p = 0.01$ and $a/\Lambda = 0.01$ (finite-diameter-particle approach). As a reference also $D_{v_0,\text{eff}}/D_{v_0,\text{eff}}(\infty)$ is shown.

The results are given in Figs. 1–8. In Figs. 1 (point-particle approximation) and 2 (finite-diameter-particle approximation), $D_{u,\text{eff}}(t)/D_{v_0,\text{eff}}(\infty)$ is plotted as function of $\nu t/\Lambda^2$ for several values of the particle volume fraction ϕ, for a ratio of the fluid density and the particle density $\rho_f/\rho_p = 0.01$ and for a ratio of the particle diameter and the turbulence integral length scale $a/\Lambda = 0.01$. (When the density ratio is 0.01 and the volume fraction is 0.1, the mass loading will be 10 and the particle phase will contain most of the momentum budget.) In Figs. 3 (point-particle approximation) and 4 (finite-diameter-particle approximation), $D_{u,\text{eff}}(t)/D_{v_0,\text{eff}}(\infty)$ is given as function of $\nu t/\Lambda^2$ for a number of values of ρ_f/ρ_p, and for $\phi = 0.1$ and $a/\Lambda = 0.01$. In Figs. 5 (point-particle approximation) and 6 (finite-diameter-particle approximation), $D_{u,\text{eff}}(t)/D_{v_0,\text{eff}}(\infty)$ is given as function of $\nu t/\Lambda^2$ for several values of a/Λ, and for $\phi = 0.1$ and $\rho_f/\rho_p = 0.01$. Finally in Figs. 7 (point-particle approximation) and 8 (finite-diameter-particle approximation), $\Delta(t)\nu/\Lambda^2 D_{v_0,\text{eff}}(\infty)$ is shown as function of $\nu t/\Lambda^2$ for two values of the volume fraction ϕ, and for $\rho_f/\rho_p = 0.01$ and $a/\Lambda = 0.01$.

From all the figures it is clear, that the particles have a very significant influence on the turbulent diffusion coefficient and on the mean square displacement in a suspension. Figs. 1 and 2 show that with increasing particle volume concentration the turbulent diffusion coefficient for the particles decreases considerably. As expected the long-term values of the diffusion coefficients $D_{u,\text{eff}}(\infty)$ and $D_{v_0,\text{eff}}(\infty)$ for particles and fluid become equal in the point-particle approximation. This is not the case for the finite-diameter-particle approximation. As can be seen from a comparison between Figs. 1 and 2, there is significant difference between the two approximations. So as already remarked by Ooms and Jansen (2000) one has to be careful with using a point-particle approximation.

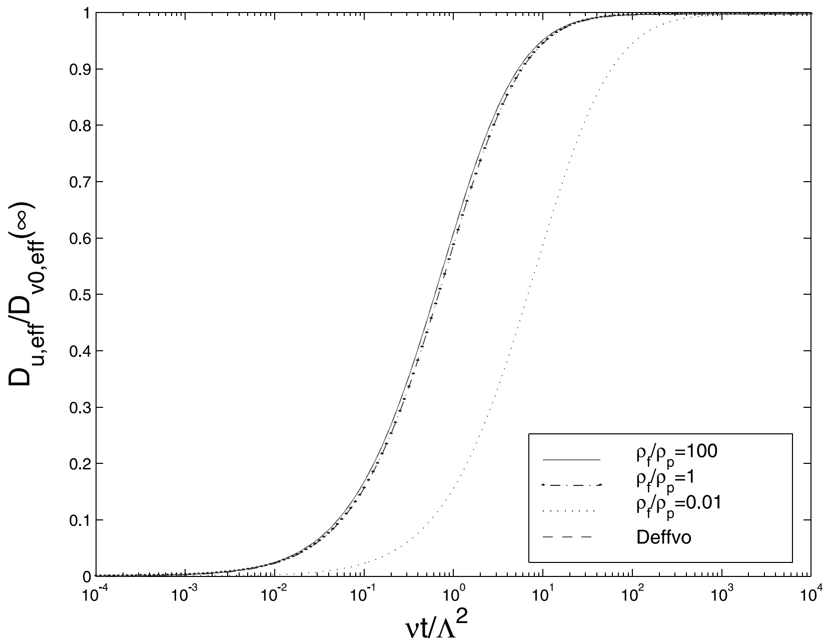

Fig. 3. Effective dimensionless diffusion coefficient for the particles as function of dimensionless time for several values of the density ratio ρ_f/ρ_p, and for $\phi = 0.1$ and $a/\Lambda = 0.01$ (point-particle approach). As a reference also $D_{v_0,\mathrm{eff}}/D_{v_0,\mathrm{eff}}(\infty)$ is shown.

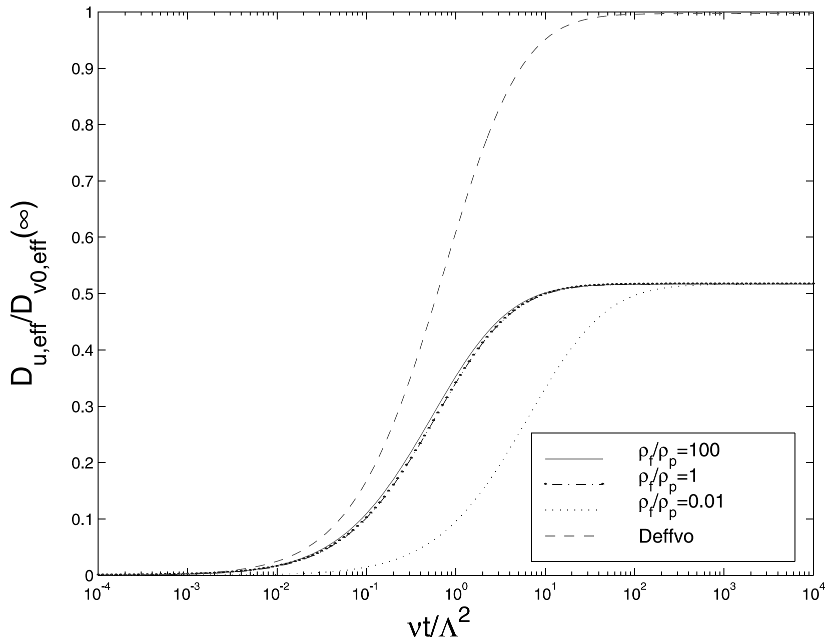

Fig. 4. Effective dimensionless diffusion coefficient for the particles as function of dimensionless time for several values of the density ratio ρ_f/ρ_p, and for $\phi = 0.1$ and $a/\Lambda = 0.01$ (finite-diameter-particle approach). As a reference also $D_{v_0,\mathrm{eff}}/D_{v_0,\mathrm{eff}}(\infty)$ is shown.

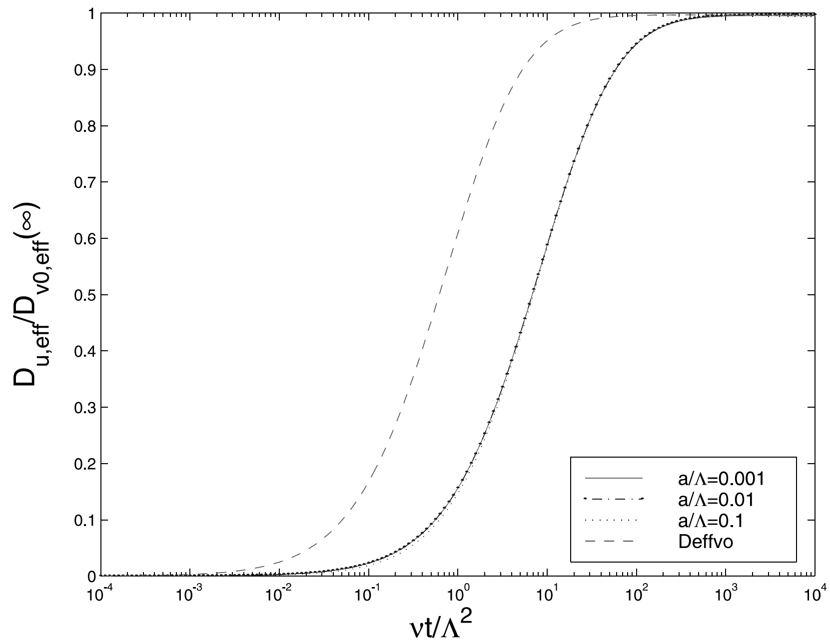

Fig. 5. Effective dimensionless diffusion coefficient for the particles as function of dimensionless time for several values of the length scale ratio a/Λ, and for $\phi = 0.1$ and $\rho_f/\rho_p = 0.01$ (point-particle approach). As a reference also $D_{v_0,\mathrm{eff}}/D_{v_0,\mathrm{eff}}(\infty)$ is shown.

In Figs. 3 and 4, the influence of the density ratio of fluid and particles on the turbulent diffusion coefficient is demonstrated. The heavier the particles are, the more pronounced is the damping influence on the diffusion coefficient. Again the results for the two approximations (point-particle versus finite-diameter-particle) is very significant. Moreover, as expected, the long-term values of the diffusion coefficients are independent of the density ratio.

In Figs. 5 and 6, the influence of the ratio of the particle size and the integral length scale of turbulence on the turbulent diffusion coefficient can be seen. It is evident that according to the theory this ratio has no significant influence on the diffusion coefficient. (As long as the ratio of the particle diameter and the integral length scale is smaller than about 0.1. For larger values of this ratio the particles will generate turbulence in their wakes. This phenomenon is not incorporated in the physical model for the dynamics of a suspension.) This negligible influence of the ratio of particle diameter and turbulent length scale was already found by Ooms and Jansen (2000) in their calculation for the turbulence intensity in a suspension.

Finally in Figs. 7 and 8, the influence of (small and heavy) particles on the mean square displacement is shown. It is clear that this influence at a particle volume fraction of 0.1 is considerable.

We have also studied the influence of the hydrodynamic interaction between the particles on the turbulent diffusion coefficient and on the mean square displacement. This was done by using the point-particle version of the theoretical model (for the dynamics of the suspension) with and without hydrodynamic interaction. It appeared that the influence of the hydrodynamic intercation is negligible. So no further results about this part of the study are given.

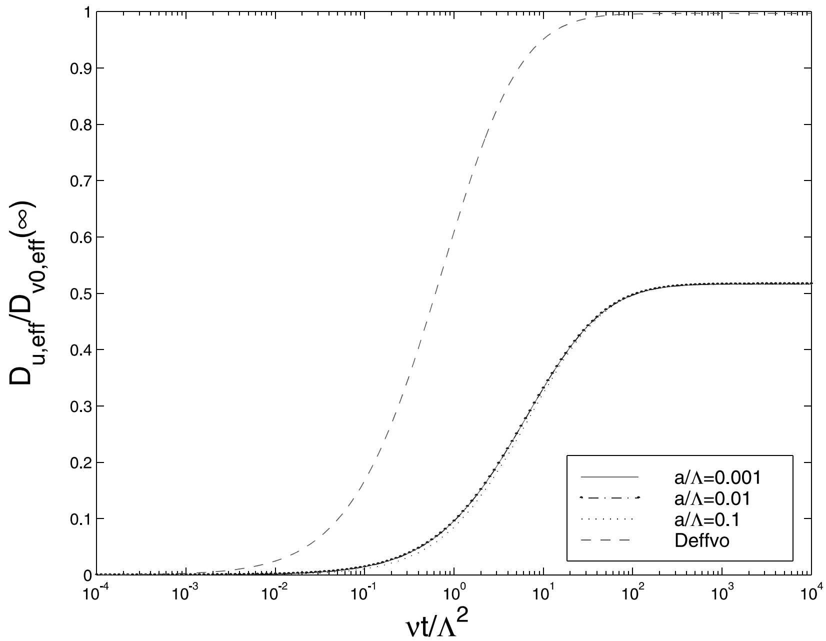

Fig. 6. Effective dimensionless diffusion coefficient for the particles as function of dimensionless time for several values of the length scale ratio a/Λ, and for $\phi = 0.1$ and $\rho_f/\rho_p = 0.01$ (finite-diameter-particle approach). As a reference also $D_{v_0,\mathrm{eff}}/D_{v_0,\mathrm{eff}}(\infty)$ is shown.

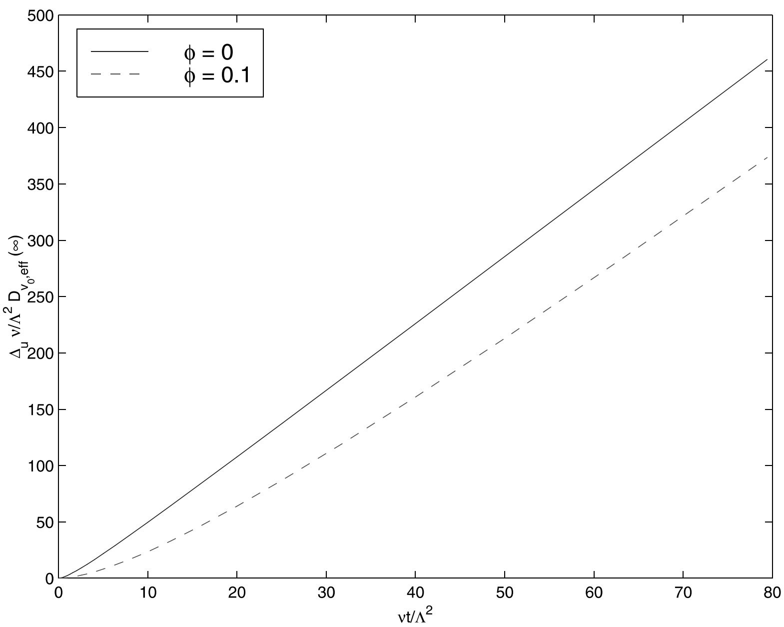

Fig. 7. Dimensionless mean square displacement as function of dimensionless time for two values of the volume fraction ϕ, and for $a/\Lambda = 0.01$ and for $\rho_f/\rho_p = 0.01$ (point-particle approach).

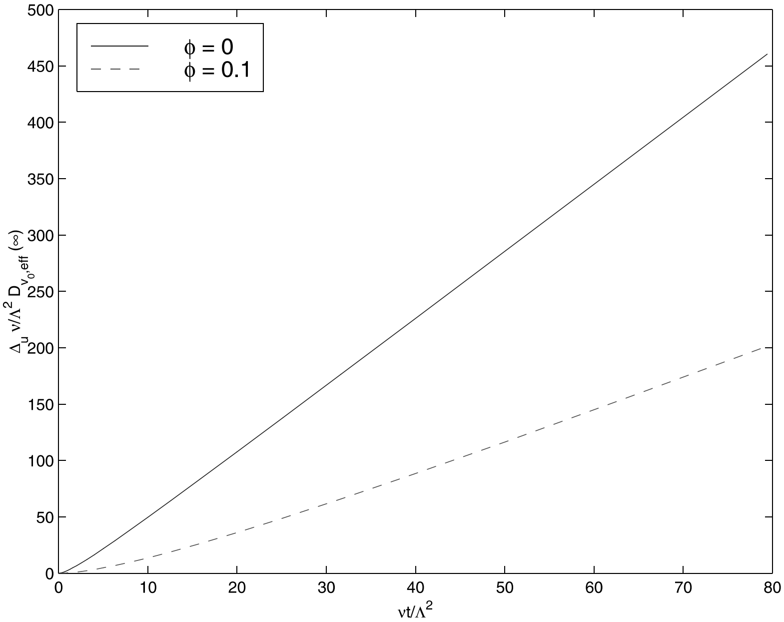

Fig. 8. Dimensionless mean square displacement as function of dimensionless time for two values of the volume fraction ϕ, and for $a/\Lambda = 0.01$ and for $\rho_f/\rho_p = 0.01$ (finite-diameter-particle approach).

5. Conclusions

It has been shown that the particles in a turbulently flowing fluid–particles suspension can have an important influence on the turbulent diffusion in the suspension. At a particle volume fraction of 0.1 and at a ratio of fluid density and particle density of 0.01 the turbulent diffusion coefficient for the particles in the suspension and the mean square displacement of a cloud of (marked) particles is significantly reduced. This reduction increases with increasing mass density of the particles. The influence of the particle diameter (when smaller than the integral length scale of the turbulence) was found to be unimportant. The differences between the results for the turbulent diffusion coefficient and for the mean square displacement calculated with the point-particle version and the finite-diameter-particle version of the theoretical model (for the dynamics of the suspension) are large. The effect of the hydrodynamic interaction between the particles is up to a volume fraction of 0.1 still negligible.

Ooms and Jansen (2000) have derived similar conclusions for the reduction of the turbulence intensity of a suspension. At the moment experiments are in preparation to check the calculated results for the turbulence intensity, the turbulent diffusion coefficient and for the mean square displacement.

Acknowledgement

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Appendix A

Consider a stationary, homogeneous and isotropic turbulent flow field without particles. The velocity field and pressure field are, respectively, given by $\mathbf{v}_0$ and p_0. In order for the flow field to remain stationary, homogeneous and isotropic an external (stirring) force field $\mathbf{F}_0$ acts on the fluid. N particles are then added to the fluid, keeping the force field $\mathbf{F}_0$. To calculate the new velocity field and pressure field the method of induced forces can be employed. In this method, the two-phase system is described by a velocity field $\mathbf{v}$, called the suspension velocity, which is identical to the fluid velocity in the part of space occupied by the fluid, and identical to the solid body motion of the particles in the part of space occupied by the particles. The effect of the stick boundary condition at the surfaces of the particles is represented by the force $\mathbf{F}$ acting on the fluid. The relation between the suspension velocity $\mathbf{v}$ and the unperturbed (without particles) fluid velocity $\mathbf{v}_0$ can formally be written in the following way (after Fourier transformation with respect to time):

$$\mathbf{v}_\omega(1,\ldots,N;\mathbf{r}) = \int \mathrm{d}\mathbf{r}'\mathbf{K}_\omega(\mathbf{r},\mathbf{r}';1,\ldots,N)\cdot\mathbf{v}_{0,\omega}(\mathbf{r}') \tag{A.1}$$

or, briefly,

$$\mathbf{v}_\omega(1,\ldots,N) = \mathbf{K}_\omega(1,\ldots,N)\cdot\mathbf{v}_{0,\omega}, \tag{A.2}$$

ω is the frequency. Similarly, the relation between the force $\mathbf{F}$ and $\mathbf{v}_0$ can formally be written as

$$\mathbf{F}_\omega(1,\ldots,N) = \mathbf{A}_\omega(1,\ldots,N)\cdot\mathbf{v}_{0,\omega}. \tag{A.3}$$

(The calculation of the operators $\mathbf{K}_\omega$ and $\mathbf{A}_\omega$ will be discussed.) The probability distribution of the particles in the fluid flow field is given by $P(1,\ldots,N)$. Ensemble averaging of Eqs. (A.2) and (A.3) yields the average relations

$$\langle\mathbf{v}_\omega\rangle = \int\cdots\int \mathrm{d}\mathbf{R}_1\cdots\mathrm{d}\mathbf{R}_N\,P(1,\ldots,N)\,\mathbf{K}_\omega(1,\ldots,N)\cdot\mathbf{v}_{0,\omega}, \tag{A.4}$$

$$\langle\mathbf{F}_\omega\rangle = \int\cdots\int \mathrm{d}\mathbf{R}_1\cdots\mathrm{d}\mathbf{R}_N\,P(1,\ldots,N)\,\mathbf{A}_\omega(1,\ldots,N)\cdot\mathbf{v}_{0,\omega}, \tag{A.5}$$

$\langle\mathbf{v}_\omega\rangle$ is the mean suspension velocity. In order to separate the fluid and particle motion, it is convenient to introduce the mean fluid velocity $\mathbf{v}_{\mathrm{f}\,\omega}$ and the mean particle velocity $\mathbf{v}_{\mathrm{p}\,\omega}$. These velocities are related to the mean suspension velocity by

$$\langle\mathbf{v}_\omega\rangle = (1-\phi)\mathbf{v}_{\mathrm{f}\,\omega} + \phi\mathbf{v}_{\mathrm{p}\,\omega}, \tag{A.6}$$

in which ϕ is the volume fraction of the particles. The plan is to eliminate $\mathbf{v}_{0,\omega}$ from (A.4) and (A.5), and to derive the relation between $\langle\mathbf{F}_\omega\rangle$ and $\langle\mathbf{v}_\omega\rangle$:

$$\langle\mathbf{F}_\omega\rangle = \chi^*_\omega\cdot\langle\mathbf{v}_\omega\rangle \tag{A.7}$$

with χ^*_ω the effective friction operator, which has to be determined. To that purpose we first introduce cluster expansions for $\mathbf{K}_\omega(1,\ldots,N)$ and $\mathbf{A}_\omega(1,\ldots,N)$ in relations (A.4) and (A.5). These relations then become expansions with their terms ordered with respect to the number of particles involved. For (A.4) we introduce the cluster operators $\mathbf{L}_\omega$ defined in the following way:

$$\begin{aligned}
\mathbf{K}_\omega(\Phi) &= \mathbf{L}_\omega(\Phi) = \mathbf{1},\\
\mathbf{K}_\omega(1) &= \mathbf{L}_\omega(1) + \mathbf{L}_\omega(\Phi),\\
\mathbf{K}(1,2) &= \mathbf{L}_\omega(1,2) + \mathbf{L}_\omega(1) + \mathbf{L}_\omega(2) + \mathbf{L}_\omega(\Phi),\\
&\;\vdots
\end{aligned}$$

Φ is the empty set. For (A.5) we choose particle 1 as reference particle, and introduce so-called rooted cluster operators $\mathbf{M}_\omega$, defined as

$$\begin{aligned}
\mathbf{A}_\omega(1) &= \mathbf{M}_\omega(1),\\
\mathbf{A}(1,2) &= \mathbf{M}_\omega(1;2) + \mathbf{M}_\omega(1),\\
&\;\vdots
\end{aligned}$$

Substitution in (A.4) and (A.5) gives after some calculations

$$\langle \mathbf{v}_\omega \rangle = \sum_{s=0}^{N} \frac{1}{s!} \int \cdots \int \mathrm{d}\mathbf{R}_1 \cdots \mathrm{d}\mathbf{R}_s\, n(1,\ldots,s)\, \mathbf{L}_\omega(1,\ldots,s) \cdot \mathbf{v}_{0,\omega}, \tag{A.8}$$

$$\langle \mathbf{F}_\omega \rangle = \sum_{s=1}^{N} \frac{1}{(s-1)!} \int \cdots \int \mathrm{d}\mathbf{R}_1 \cdots \mathrm{d}\mathbf{R}_s\, n(1,\ldots,s)\, \mathbf{M}_\omega(1;2,\ldots,s) \cdot \mathbf{v}_{0,\omega}, \tag{A.9}$$

in which $n(1,\ldots,s)$ is the partial probability distribution function defined as

$$n(1,\ldots,s) = \frac{N!}{(N-s)!} \int \cdots \int \mathrm{d}\mathbf{R}_{s+1} \cdots \mathrm{d}\mathbf{R}_N\, P(1,\ldots,N). \tag{A.10}$$

Elimination of $\mathbf{v}_{0,\omega}$ from (A.8) and (A.9) gives the following first two terms in the expansion for the effective friction operator χ^*_ω:

$$\begin{aligned}
\chi^*_\omega = &\int \mathrm{d}\mathbf{R}_1\, n(1)\, \mathbf{M}_\omega(1)\\
&+ \int\int \mathrm{d}\mathbf{R}_1\, \mathrm{d}\mathbf{R}_2 [n(1,2)\, \mathbf{M}_\omega(1;2) - n(1)\, n(2)\, \mathbf{M}_\omega(1) \cdot \mathbf{L}_\omega(2)].
\end{aligned} \tag{A.11}$$

$\mathbf{M}_\omega(1)$, $\mathbf{M}_\omega(1;2)$ and $\mathbf{L}_\omega(2)$ can be calculated by solving the single-particle- and two-particle flow problem. Felderhof and Ooms (1989, 1990) have done that in point-particle approximation using Stokes' resistance law for the particles. Felderhof and Jansen (1991) have solved the single-particle flow problem for the finite-diameter-particle case. They take the full force field exerted by the particle on the fluid into account (that is all force multi-pole moments, not only the first). Their calculation method can handle the two-particle flow problem in principle, but the calculations are very tedious and computationally intensive. Eq. (A.11) shows how the particle size and volume fraction effects are included in the calculation. Also for the point-particle approximation the particle size is included via the probability distribution function.

Substitution of Eq. (A.7) in the equation of motion gives (after spatial Fourier transformation) for the finite-diameter-particle case

$$\left[\eta\left(\alpha^2 + q^2\right) - \chi^{\mathrm{T}}(q, \omega)\right]\langle \mathbf{v}_{\mathbf{q}\omega}\rangle = (1 - \phi)\mathbf{F}^{\mathrm{T}}_{0\mathbf{q}\omega}, \tag{A.12}$$

which specifies the mean suspension velocity $\langle \mathbf{v}_{\mathbf{q}\omega}\rangle$ in response to the transversal component of the force $\mathbf{F}^{\mathrm{T}}_{0\mathbf{q}\omega}$. η is the dynamic viscosity of the fluid, q the wave number and $\mathbf{q}$ the wave vector. α is given by

$$\alpha = \left(\frac{-\mathrm{i}\omega\rho_{\mathrm{f}}}{\eta}\right)^{1/2}, \tag{A.13}$$

where ρ_{f} is the fluid density. The function $\chi^{\mathrm{T}}(q, \omega)$ is the transversal component of the effective friction operator defined by

$$\chi^{*}_{\omega} = \chi^{\mathrm{L}}(q, \omega)\hat{\mathbf{q}}\hat{\mathbf{q}} + \chi^{\mathrm{T}}(q, \omega)(\mathbf{1} - \hat{\mathbf{q}}\hat{\mathbf{q}}), \tag{A.14}$$

in which $\hat{\mathbf{q}}$ is the unit vector in the direction of $\mathbf{q}$. For an incompressible fluid the pressure term does not occur in the equation of motion for the transversal component of the force. It is in the equation for the lateral component.

$\mathbf{F}^{\mathrm{T}}_{0\mathbf{q}\omega}$ can also be related to the mean unperturbed (without particles) fluid velocity field $\mathbf{v}_{0\mathbf{q}\omega}$ by means of the following expression:

$$\left[\eta\left(\alpha^2 + q^2\right)\right]\mathbf{v}_{0\mathbf{q}\omega} = \mathbf{F}^{\mathrm{T}}_{0\mathbf{q}\omega}. \tag{A.15}$$

Substitution of (A.15) in (A.12) yields the relation between the suspension velocity and the unperturbed fluid velocity:

$$\langle \mathbf{v}_{\mathbf{q}\omega}\rangle = (1 - \phi)H(q, \omega)\mathbf{v}_{0\mathbf{q}\omega}, \tag{A.16}$$

where the function $H(q, \omega)$ is given by

$$H(q, \omega) = \frac{\eta(\alpha^2 + q^2)}{\eta(\alpha^2 + q^2) - \chi^{\mathrm{T}}(q, \omega)}. \tag{A.17}$$

Straightforward calculations yield similar relations for the mean particle velocity $\mathbf{v}_{\mathrm{p}\,\mathbf{q}\omega}$ and the mean fluid velocity $\mathbf{v}_{\mathrm{f}\,\mathbf{q}\omega}$ for the finite-diameter-particle case in terms of $\mathbf{v}_{0\mathbf{q}\omega}$ and $p_{0\mathbf{q}\omega}$:

$$\mathbf{v}_{\mathrm{p}\,\mathbf{q}\omega} = \left\{\left[(1 - \phi)\boldsymbol{\Gamma}^{\mathrm{T}}(q, \omega)\right]H(q, \omega) + \boldsymbol{\Gamma}^{\mathrm{T}}_{F}(q, \omega)\eta\left(\alpha^2 + q^2\right)\right\}\mathbf{v}_{0\mathbf{q}\omega} + \boldsymbol{\Gamma}^{\mathrm{L}}_{F}(q, \omega)\,\mathrm{i}\,\mathbf{q}p_{0\mathbf{q}\omega}, \tag{A.18}$$

$$\begin{aligned}\mathbf{v}_{\mathrm{f}\,\mathbf{q}\omega} = {} & \left\{\left[1 - \phi\boldsymbol{\Gamma}^{\mathrm{T}}(q, \omega)\right]H(q, \omega) - \frac{\phi}{1 - \phi}\boldsymbol{\Gamma}^{\mathrm{T}}_{F}(q, \omega)\eta\left(\alpha^2 + q^2\right)\right\}\mathbf{v}_{0\mathbf{q}\omega} \\ & - \frac{\phi}{1 - \phi}\boldsymbol{\Gamma}^{\mathrm{L}}_{F}(q, \omega)\,\mathrm{i}\,\mathbf{q}p_{0\mathbf{q}\omega}.\end{aligned} \tag{A.19}$$

The functions $\chi^{\mathrm{T}}(q, \omega)$, $\boldsymbol{\Gamma}^{\mathrm{T}}(q, \omega)$, $\boldsymbol{\Gamma}^{\mathrm{T}}_{F}(q, \omega)$ and $\boldsymbol{\Gamma}^{\mathrm{L}}_{F}(q, \omega)$ are given in Felderhof and Jansen (1991).

Within the point-particle approach the force density $\mathbf{F}^{\mathrm{T}}_{0\mathbf{q}\omega}$ acts in the entire space, whereas in the finite-diameter-particle approach its action is restricted to the space occupied by the fluid. Accordingly, Eq. (A.12) differs for the point-particle case by the fact that the factor $(1 - \phi)$ in the right-hand side is absent. The equivalent of Eq. (A.16) is therefore given by

$$\langle \mathbf{v}_{\mathbf{q}\omega}\rangle = H(q, \omega)\mathbf{v}_{0\mathbf{q}\omega}, \tag{A.20}$$

where the function $H(q, \omega)$ is as in (A.17). Within the point-particle approach the mean fluid velocity $\mathbf{v}_{\mathrm{f}\,\mathbf{q}\omega}$ is identical to the suspension velocity $\langle \mathbf{v}_{\mathbf{q}\omega}\rangle$

$$\mathbf{v}_{\mathrm{f}\,\mathbf{q}\omega} = \langle \mathbf{v}_{\mathbf{q}\omega} \rangle = H(q,\omega)\mathbf{v}_{0\mathbf{q}\omega}, \tag{A.21}$$

while the mean particle velocity field $\mathbf{v}_{\mathrm{p}\,\mathbf{q}\omega}$ can be expressed as

$$\mathbf{v}_{\mathrm{p}\,\mathbf{q}\omega} = \boldsymbol{\Gamma}^{\mathrm{T}}(q,\omega)\langle \mathbf{v}_{\mathbf{q}\omega} \rangle = \boldsymbol{\Gamma}^{\mathrm{T}}(q,\omega)H(q,\omega)\mathbf{v}_{0\mathbf{q}\omega}. \tag{A.22}$$

$\boldsymbol{\Gamma}^{\mathrm{T}}(q,\omega)$ and $\chi^{\mathrm{T}}(q,\omega)$ for the point-particle approach are different from their expressions for the finite-diameter-particle approach, as in the point-particle approach the hydrodynamic interactions between the particles (in two-particle approximation) are taken into account. They are specified in Felderhof and Ooms (1989, 1990).

Given the unperturbed (fluid without particles) spectral density $S_0(q,\omega)$, the perturbed (fluid with particles) fluid spectral density $S_{\mathrm{f}}(q,\omega)$ and the particle spectral density $S_{\mathrm{p}}(q,\omega)$ can be derived using Eqs. (A.19) and (A.18) for the fluid velocity field and the particle velocity field, respectively, in case of the finite-diameter-particle approach. This yields after straightforward calculations:

$$\begin{aligned} S_{\mathrm{f}}(q,\omega) = \Bigg\{ &\left| [1-\phi\boldsymbol{\Gamma}^{\mathrm{T}}(q,\omega)]H(q,\omega) - \frac{\phi}{1-\phi}\tilde{\boldsymbol{\Gamma}}^{\mathrm{T}}_{F}(q,\omega) \right|^2 \\ &+ \frac{1}{2}\left(\frac{\phi}{1-\phi}\right)^2 \left|\tilde{\boldsymbol{\Gamma}}^{\mathrm{L}}_{F}(q,\omega)\right|^2 \Bigg\} S_0(q,\omega), \end{aligned} \tag{A.23}$$

$$S_{\mathrm{p}}(q,\omega) = \left\{ \left| \boldsymbol{\Gamma}^{\mathrm{T}}(q,\omega)(1-\phi)H(q,\omega) + \tilde{\boldsymbol{\Gamma}}^{\mathrm{T}}_{F}(q,\omega) \right|^2 + \frac{1}{2}\left|\tilde{\boldsymbol{\Gamma}}^{\mathrm{L}}_{F}(q,\omega)\right|^2 \right\} S_0(q,\omega), \tag{A.24}$$

where

$$\tilde{\boldsymbol{\Gamma}}^{\mathrm{T}}_{F}(q,\omega) = \eta(\alpha^2+q^2)\boldsymbol{\Gamma}^{\mathrm{T}}_{F}(q,\omega), \quad \tilde{\boldsymbol{\Gamma}}^{\mathrm{L}}_{F}(q,\omega) = \eta(\alpha^2+q^2)\boldsymbol{\Gamma}^{\mathrm{L}}_{F}(q,\omega). \tag{A.25}$$

With Eqs. (A.23) and (A.24) the relevant spectra can be calculated from the spectrum of the unperturbed fluid in the finite-diameter-particle approach.

For the point-particle approach it can be shown, that these equations can be written in the following forms:

$$S_{\mathrm{f}} = |H(q,\omega)|^2 S_0(q,\omega), \tag{A.26}$$

$$S_{\mathrm{p}} = \left|\boldsymbol{\Gamma}^{\mathrm{T}}(q,\omega)\right|^2 |H(q,\omega)|^2 S_0(q,\omega). \tag{A.27}$$

Appendix B

We start from

$$\frac{\partial n_1}{\partial t} - \mathbf{A}_0 n_1 - \mathbf{A}_1 n_1 = s. \tag{B.1}$$

Ensemble averaging yields

$$\frac{\partial \langle n_1 \rangle}{\partial t} - \mathbf{A}_0\langle n_1 \rangle - \langle \mathbf{A}_1 n_1 \rangle = s. \tag{B.2}$$

We need an expression for $\langle \mathbf{A}_1 n_1 \rangle$. Eq. (B.1) can be written as

$$\left[\frac{\partial}{\partial t} - \mathbf{A}_0 - \mathbf{A}_1\right] n_1 = s \tag{B.3}$$

or

$$n_1 = \mathbf{G}s \tag{B.4}$$

with

$$\mathbf{G} = \left[\frac{\partial}{\partial t} - \mathbf{A}_0 - \mathbf{A}_1\right]^{-1}. \tag{B.5}$$

Eq. (B.4) gives after ensemble averaging

$$\langle n_1 \rangle = \langle \mathbf{G} \rangle s, \tag{B.6}$$

so

$$s = \langle \mathbf{G} \rangle^{-1} \langle n_1 \rangle. \tag{B.7}$$

$\langle \mathbf{A}_1 n_1 \rangle$ can therefore be written as

$$\langle \mathbf{A}_1 n_1 \rangle = \langle \mathbf{A}_1 G \rangle s \tag{B.8}$$

or

$$\langle \mathbf{A}_1 n_1 \rangle = \langle \mathbf{A}_1 \mathbf{G} \rangle \langle \mathbf{G} \rangle^{-1} \langle n_1 \rangle. \tag{B.9}$$

This is an exact relation for $\langle \mathbf{A}_1 n_1 \rangle$, but it is too complex for calculations. We will therefore derive an approximation. Let us first introduce $\mathbf{G}_0$,

$$\mathbf{G}_0 = \left[\frac{\partial}{\partial t} - \mathbf{A}_0\right]^{-1}, \tag{B.10}$$

and prove that

$$\mathbf{G} = \mathbf{G}_0 + \mathbf{G}_0\mathbf{A}_1\mathbf{G}_0 + \mathrm{O}\left(\mathbf{A}_1^2\right). \tag{B.11}$$

To prove that, we rewrite Eq. (B.5) in the following form

$$\mathbf{G}^{-1} = \mathbf{G}_0^{-1} - \mathbf{A}_1, \tag{B.12}$$

and prove that $\mathbf{G}^{-1}\mathbf{G} = \mathbf{G}\mathbf{G}^{-1} = \mathbf{1} + \mathrm{O}(\mathbf{A}_1^2)$, where $\mathbf{G}^{-1}$ is taken from (B.12) and $\mathbf{G}$ from (B.11).

Proof:

$$\begin{aligned}\mathbf{G}^{-1}\mathbf{G} &= \left[\mathbf{G}_0^{-1} - \mathbf{A}_1\right]\left[\mathbf{G}_0 + \mathbf{G}_0\mathbf{A}_1\mathbf{G}_0 + \mathrm{O}\left(\mathbf{A}_1^2\right)\right] \\ &= \mathbf{1} + \mathbf{A}_1\mathbf{G}_0 - \mathbf{A}_1\mathbf{G}_0 + \mathrm{O}\left(\mathbf{A}_1^2\right) = \mathbf{1} + \mathrm{O}\left(\mathbf{A}_1^2\right),\end{aligned} \tag{B.13}$$

$$\begin{aligned}\mathbf{G}\mathbf{G}^{-1} &= \left[\mathbf{G}_0 + \mathbf{G}_0\mathbf{A}_1\mathbf{G}_0 + \mathrm{O}\left(\mathbf{A}_1^2\right)\right]\left[\mathbf{G}_0^{-1} - \mathbf{A}_1\right] \\ &= \mathbf{1} - \mathbf{G}_0\mathbf{A}_1 + \mathbf{G}_0\mathbf{A}_1 + \mathrm{O}\left(\mathbf{A}_1^2\right) = \mathbf{1} + \mathrm{O}\left(\mathbf{A}_1^2\right).\end{aligned} \tag{B.14}$$

Let us finally prove that

$$\langle \mathbf{A}_1\mathbf{G} \rangle \left\langle \mathbf{G}^{-1} \right\rangle = \langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1 \rangle + \mathrm{O}\left(\mathbf{A}_1^3\right) \tag{B.15}$$

or that

$$\langle \mathbf{A}_1\mathbf{G}\rangle = \langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\langle \mathbf{G}\rangle + \mathrm{O}\big(\mathbf{A}_1^3\big). \tag{B.16}$$

To that purpose we calculate $\langle \mathbf{A}_1\mathbf{G}\rangle$ and $\langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\langle \mathbf{G}\rangle$, using approximation (B.11) and using $\langle \mathbf{A}_1\rangle = 0$, and compare the results:

$$\begin{aligned}\langle \mathbf{A}_1\mathbf{G}\rangle &= \big\langle \mathbf{A}_1\mathbf{G}_0 + \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\mathbf{G}_0 + \mathrm{O}\big(\mathbf{A}_1^3\big)\big\rangle = \langle \mathbf{A}_1\rangle\mathbf{G}_0 + \langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\mathbf{G}_0 + \big\langle \mathrm{O}\big(\mathbf{A}_1^3\big)\big\rangle \\ &= \langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\mathbf{G}_0 + \big\langle \mathrm{O}\big(\mathbf{A}_1^3\big)\big\rangle,\end{aligned} \tag{B.17}$$

$$\langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\langle \mathbf{G}\rangle = \langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\langle \mathbf{G}_0 + \mathrm{O}(\mathbf{A}_1)\rangle = \langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\mathbf{G}_0 + \mathrm{O}\big(\mathbf{A}_1^3\big). \tag{B.18}$$

From (B.17) and (B.18) follows

$$\langle \mathbf{A}_1\mathbf{G}\rangle = \langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\langle \mathbf{G}\rangle + \big\langle \mathrm{O}\big(\mathbf{A}_1^3\big)\big\rangle. \tag{B.19}$$

Substitution of (B.19) and (B.9) in (B.2) gives within $\mathrm{O}(\mathbf{A}_1^3)$ approximation the following equation:

$$\left[\frac{\partial}{\partial t} - \mathbf{A}_0 - \langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\right]\langle n_1\rangle = s. \tag{B.20}$$

Appendix C

We start from the following equation:

$$\left[\frac{\partial}{\partial t} - \mathbf{A}_0 - \langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\right]\langle n_1\rangle = s. \tag{C.1}$$

We assume the limit of vanishing D_m; so

$$\mathbf{A}_0 = 0 \tag{C.2}$$

$$\mathbf{G}_0 = \left[\frac{\partial}{\partial t}\right]^{-1} = \int_{-\infty}^{t} \mathrm{d}t. \tag{C.3}$$

Let us now first calculate $\langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\langle n_1\rangle$:

$$\langle \mathbf{A}_1\mathbf{G}_0\mathbf{A}_1\rangle\langle n_1\rangle = \left\langle \mathbf{A}_1(t)\int_{-\infty}^{t} \mathbf{A}_1(t')\right\rangle\langle n_1(t')\rangle\,\mathrm{d}t' = \int_{-\infty}^{t}\langle \mathbf{A}_1(t)\mathbf{A}_1(t')\rangle\langle n_1(t')\rangle\,\mathrm{d}t'. \tag{C.4}$$

For $\mathbf{A}_1(t)\mathbf{A}_1(t')$ we write with $\mathbf{A}_1(\mathbf{r},\mathbf{r}') = \nabla\cdot\mathbf{u}(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}')$

$$\mathbf{A}_1(t)\mathbf{A}_1(t') = \int \nabla\cdot\mathbf{u}(\mathbf{r},t)\delta(\mathbf{r}-\mathbf{r}'')\nabla''\cdot\mathbf{u}(\mathbf{r}'',t')\delta(\mathbf{r}''-\mathbf{r}')\delta\mathbf{r}''. \tag{C.5}$$

So

$$\begin{aligned}\langle \mathbf{A}_1(t)\mathbf{A}_1(t')\rangle &= \int \nabla\cdot\delta(\mathbf{r}-\mathbf{r}'')\nabla''\cdot\delta(\mathbf{r}''-\mathbf{r}')\langle \mathbf{u}(\mathbf{r},t)\mathbf{u}(\mathbf{r}'',t')\rangle\,\mathrm{d}\mathbf{r}'' \\ &= \int \nabla\cdot\delta(\mathbf{r}-\mathbf{r}'')\nabla''\cdot\delta(\mathbf{r}''-\mathbf{r}')\boldsymbol{\Gamma}_u(\mathbf{r}-\mathbf{r}'',t-t')\,\mathrm{d}\mathbf{r}''\end{aligned} \tag{C.6}$$

with $\mathbf{\Gamma}_u(\mathbf{r}-\mathbf{r}'', t-t')$ the correlation function of the turbulent particle velocity field. We assume homogeneous, isotropic turbulence. The correlation function then has the following form:

$$\mathbf{\Gamma}_u(\mathbf{r},t) = \frac{1}{3}\langle \mathbf{u}^2\rangle [f_u(\mathbf{r},t)\hat{r}\hat{r} + g_u(\mathbf{r},t)(\mathbf{1}-\hat{r}\hat{r})], \tag{C.7}$$

where the scalar functions have the following form:

$$f_u(0,t) = g_u(0,t) \quad \text{and} \quad f_u(0,0) = 1. \tag{C.8}$$

From Eq. (C.6) we find

$$\begin{aligned}\langle \mathbf{A}_1(t)\mathbf{A}_1(t')\rangle\langle n_1\rangle &= \int \nabla\cdot\delta(\mathbf{r}-\mathbf{r}'')\nabla''\cdot\delta(\mathbf{r}''-\mathbf{r}')\mathbf{\Gamma}_u(\mathbf{r}-\mathbf{r}'',t-t')\langle n_1(\mathbf{r}',t')\rangle\,\mathrm{d}\mathbf{r}'\,\mathrm{d}\mathbf{r}'' \\ &= \nabla\cdot\int \delta(\mathbf{r}-\mathbf{r}'')\nabla''\cdot\mathbf{\Gamma}(\mathbf{r}-\mathbf{r}'',t-t')\langle n_1(\mathbf{r}'',t')\rangle\,\mathrm{d}\mathbf{r}'' \\ &= \nabla^2[\mathbf{\Gamma}(0,t-t')\langle n_1(\mathbf{r},t')\rangle].\end{aligned} \tag{C.9}$$

Substituting (C.7) and (C.8) yields

$$\langle \mathbf{A}_1(t)\mathbf{A}_1(t')\rangle\langle n_1(t')\rangle = \mathbf{\Gamma}_u(0,t-t')\nabla^2\langle n_1(t')\rangle. \tag{C.10}$$

$\mathbf{\Gamma}_u(0,t-t')$ in (C.10) is defined by

$$\mathbf{\Gamma}_u(0,t-t') = \mathbf{\Gamma}_u(0,t-t')\mathbf{1} \tag{C.11}$$

with

$$\mathbf{\Gamma}_u(0,t-t') = \tfrac{1}{3}\langle \mathbf{u}^2\rangle f_u(0,t-t'). \tag{C.12}$$

Substitution of (C.10), with (C.4) and (C.2), in (C.1) gives

$$\frac{\partial}{\partial t}\langle n_1(\mathbf{r},t)\rangle - \int_{-\infty}^{t} \mathbf{\Gamma}_u(0,t-t')\nabla^2\langle n_1(\mathbf{r},t')\rangle\,\mathrm{d}t' = s(\mathbf{r},t). \tag{C.13}$$

We rewrite (C.13) in terms of the following Fourier transforms:

$$n_1(\mathbf{r},t) = \int_{-\infty}^{+\infty} n_{1,\omega}(\mathbf{r})\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}\omega \tag{C.14}$$

$$s(\mathbf{r},t) = \int_{-\infty}^{+\infty} s_\omega(\mathbf{r})\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}\omega. \tag{C.15}$$

Transforming (C.13) we find

$$\begin{aligned}&-\mathrm{i}\omega\left\langle \int_{-\infty}^{+\infty} n_{1,\omega}(\mathbf{r})\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}\omega\right\rangle - \int_{-\infty}^{t} \mathbf{\Gamma}_u(0,t-t')\nabla^2\left\langle \int_{-\infty}^{+\infty} n_{1,\omega}(\mathbf{r})\,\mathrm{e}^{-\mathrm{i}\omega t'}\,\mathrm{d}\omega\right\rangle \mathrm{d}t' \\ &\quad = \int_{-\infty}^{+\infty} s_\omega(\mathbf{r})\,\mathrm{e}^{-\mathrm{i}\omega t}\,\mathrm{d}\omega\end{aligned} \tag{C.16}$$

or

$$-\mathrm{i}\omega\langle n_1(\mathbf{r})\rangle - D_u(\omega)\nabla^2\langle n_{1,\omega}(\mathbf{r})\rangle = s_\omega(\mathbf{r}) \tag{C.17}$$

with

$$D_u(\omega) = \int_0^\infty e^{i\omega t}\boldsymbol{\Gamma}_u(0,t)\,dt. \tag{C.18}$$

Appendix D

In Appendix C, we defined the correlation function $\boldsymbol{\Gamma}_u(\mathbf{r},t)$ of the turbulent particle velocity field as

$$\boldsymbol{\Gamma}_u(\mathbf{r},t) = \langle \mathbf{u}(\mathbf{r}',t')\mathbf{u}(\mathbf{r}'+\mathbf{r},t'+t)\rangle. \tag{D.1}$$

For a homogeneous and isotropic flow field it was also found that

$$\boldsymbol{\Gamma}_u(\mathbf{0},t) = \Gamma_u(0,t)\mathbf{1} \tag{D.2}$$

with

$$\Gamma_u(0,t) = \tfrac{1}{3}\langle \mathbf{u}^2\rangle f_u(0,t). \tag{D.3}$$

Fourier transformation of (D.1) with respect to time yields

$$\boldsymbol{\Gamma}_u(\mathbf{r},\omega) = \frac{1}{2\pi}\int_{-\infty}^{+\infty}\boldsymbol{\Gamma}_u(\mathbf{r},t)\,e^{i\omega t}\,dt. \tag{D.4}$$

Spatial Fourier transformation of (D.4) gives the spectral density tensor

$$\mathbf{S}_u(\mathbf{q},\omega) = \frac{1}{8\pi^3}\int \boldsymbol{\Gamma}_u(\mathbf{r},\omega)\,e^{-i\mathbf{qr}}\,d\mathbf{r}, \tag{D.5}$$

in which $\mathbf{q}$ is the wave vector. For a homogeneous and isotropic flow field the spectral density tensor can be written in the following way:

$$\mathbf{S}_u(\mathbf{q},\omega) = S_u(q,\omega)(\mathbf{1}-\hat{q}\hat{q}), \tag{D.6}$$

where q is the wave number and $\hat{q}$ a vector of unit length in the direction of the wave vector. It can be proven that the following relation exists between $\boldsymbol{\Gamma}_u(0,0)$ and $S_u(q,\omega)$:

$$\boldsymbol{\Gamma}_u(0,0) = \langle u^2\rangle = \frac{8\pi}{3}\int_{-\infty}^{+\infty}\int_0^\infty S_u(q,\omega)q^2\,dq\,d\omega. \tag{D.7}$$

It can also be shown, that

$$\boldsymbol{\Gamma}_u(0,t) = \langle u(t')u(t'+t)\rangle = \langle u^2\rangle f_u(0,t) = \frac{8\pi}{3}\int_{-\infty}^{+\infty}\int_0^\infty S_u(q,\omega)q^2\,e^{i\omega t}\,dq\,d\omega. \tag{D.8}$$

From (D.8) the following relation can be derived:

$$Re\left[\frac{1}{2\pi}\int_{-\infty}^{+\infty}\boldsymbol{\Gamma}_u(0,t)\,e^{-i\omega t}\,dt\right] = \frac{8\pi}{3}\int_0^\infty S_u(q,\omega)q^2\,dq. \tag{D.9}$$

This can be written as

$$Re\left[\frac{1}{2\pi}\int_{-\infty}^{+\infty}\boldsymbol{\Gamma}_u(0,t)\,\mathrm{e}^{\mathrm{i}\,\omega t}\,\mathrm{d}t\right]=\frac{8\pi}{3}\int_0^{\infty}S_u(q,\omega)q^2\,\mathrm{d}q \tag{D.10}$$

or, because of the symmetry of $\boldsymbol{\Gamma}_u(0,t)$,

$$Re\left[\frac{1}{\pi}\int_{0}^{\infty}\boldsymbol{\Gamma}_u(0,t)\,\mathrm{e}^{\mathrm{i}\,\omega t}\,\mathrm{d}t\right]=\frac{8\pi}{3}\int_0^{\infty}S_u(q,\omega)q^2\,\mathrm{d}q. \tag{D.11}$$

Using the definition for $D_u(\omega)$ given in Appendix C this yields

$$Re\,D_u(\omega)=\frac{8\pi^2}{3}\int_0^{\infty}S_u(q,\omega)q^2\,\mathrm{d}q. \tag{D.12}$$

Appendix E

In Appendix C the following equation was derived:

$$\frac{\partial}{\partial t}\langle n_1(\mathbf{r},t)\rangle-\int_{-\infty}^{t}\boldsymbol{\Gamma}_u(0,t-t')\nabla^2\langle n_1(\mathbf{r},t')\rangle\,\mathrm{d}t'=s(\mathbf{r},t). \tag{E.1}$$

For an initial value problem with $s=0$ this can be written as

$$\frac{\partial}{\partial t}\langle n_1(\mathbf{r},t)\rangle-\int_{0}^{t}\boldsymbol{\Gamma}_u(0,t-t')\nabla^2\langle n_1(\mathbf{r},t')\rangle\,\mathrm{d}t'=0. \tag{E.2}$$

We assume the initial value of $n_1(\mathbf{r},t)$ to be given by

$$n_1(\mathbf{r},0)=\delta(\mathbf{r}). \tag{E.3}$$

Multiplication of (E.2) by $\mathbf{r}^2$ and integration yields

$$\frac{\partial}{\partial t}\int\mathbf{r}^2\langle n_1(\mathbf{r},t)\rangle\,\mathrm{d}\mathbf{r}=\int_0^t\boldsymbol{\Gamma}_u(0,t-t')\int\mathbf{r}^2\nabla^2 n_1(\mathbf{r},t')\,\mathrm{d}\mathbf{r}\,\mathrm{d}t'. \tag{E.4}$$

Let us evaluate the integral with respect to $\mathbf{r}$ on the right-hand side of (E.4):

$$\begin{aligned}\int\mathbf{r}^2\nabla^2 n_1(\mathbf{r},t')\,\mathrm{d}\mathbf{r}&=-\int\left(\nabla\mathbf{r}^2\right)\cdot\nabla n_1(\mathbf{r},t')\,\mathrm{d}\mathbf{r}\\&=-2\int\mathbf{r}\cdot\nabla n_1(\mathbf{r},t')\,\mathrm{d}\mathbf{r}\\&=2\int(\nabla\cdot\mathbf{r})n_1(\mathbf{r},t')\,\mathrm{d}\mathbf{r}\\&=6\int n_1(\mathbf{r},t')\,\mathrm{d}\mathbf{r}=6.\end{aligned} \tag{E.5}$$

Substitution in (E.4) gives

$$\frac{\partial}{\partial t}\int\mathbf{r}^2\langle n_1(\mathbf{r},t)\rangle\,\mathrm{d}r=6\int_0^t\boldsymbol{\Gamma}_u(0,t-t')\,\mathrm{d}t'=6\int_0^t\boldsymbol{\Gamma}_u(0,\tau)\,\mathrm{d}\tau. \tag{E.6}$$

We define the effective diffusion coefficient in the following way:

$$D_{u,\mathrm{eff}} = \int_0^t \mathbf{\Gamma}_u(0,\tau)\,\mathrm{d}\tau. \tag{E.7}$$

Substitution in (E.6) gives for the mean displacement as function of time

$$\Delta(t) = \int \mathbf{r}^2 \langle n_1(\mathbf{r},t\,|\,\mathbf{0},0)\rangle\,\mathrm{d}\mathbf{r} = 6\int_0^t D_{u,\mathrm{eff}}(\tau)\,\mathrm{d}\tau. \tag{E.8}$$

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