PHYSICAL REVIEW E VOLUME 58, NUMBER 6 DECEMBER 1998

Analogies between colloidal sedimentation and turbulent convection at high Prandtl numbers

P. Tong and B. J. Ackerson
Department of Physics, Oklahoma State University, Stillwater, Oklahoma 74078

(Received 16 September 1997; revised manuscript received 11 December 1997)

A set of coarse-grained equations of motion is proposed to describe concentration and velocity fluctuations in a dilute sedimenting suspension of non-Brownian particles. With these equations, colloidal sedimentation is found to be analogous to turbulent convection at high Prandtl numbers. Using Kraichnan's mixing-length theory, scaling relations are obtained for the diffusive dissipation length δ_d, the velocity variance $\widetilde{\delta u}$, and the concentration variance $\widetilde{\delta\phi}$. The obtained scaling laws over varying particle radius a and volume fraction ϕ_0 are in excellent agreement with the recent experiment by Segrè *et al.* [Phys. Rev. Lett. **79**, 2574 (1997)]. [S1063-651X(98)50912-9]

PACS number(s): 82.70.Dd, 47.27.Te, 05.40.+j, 83.10.Pp

The study of the motions of small particles suspended in a fluid has always been an interesting subject in physics. The dynamics of the particles is determined by the statistical properties of the random forces resulting from interactions between the particle and the surrounding fluid. Brownian diffusion of small particles in a fluid at thermal equilibrium is one of the classical and best understood examples. The motion of particles in a nonequilibrium fluid, on the other hand, represents an anomalous diffusion process in which hydrodynamic dispersion coefficients might diverge [1]. Sedimentation of heavy colloidal particles under gravity through a quiescent fluid is an example of such a process. The main issue in colloidal sedimentation is to understand how hydrodynamic interactions created by the motion of many surrounding particles in the fluid affect a test particle's mean sedimentation velocity $\overline{v}$ and its variance $\widetilde{\delta v}$ at different particle concentrations [2]. Because the disturbance flow produced by a particle decays as $1/\ell$, where ℓ is the radial distance from the particle, simple theoretical calculations [3] as well as computer simulations [4] have indicated that the velocity variance $\widetilde{\delta v}$ might diverge with increasing sample size L. Experiments, however, find no dependence of $\widetilde{\delta v}$ on L [5]. Several theoretical models [1,6] have been proposed recently, aimed at resolving the divergence problem.

In a recent experiment, Segrè *et al.* [7] used the particle imaging velocimetry technique to measure the spatial correlation function, $C(\ell)=\langle \delta v(r)\delta v(r+\ell)\rangle$, of the velocity fluctuation δv in a sedimenting suspension of non-Brownian particles over a wide range of particle concentrations and sample sizes. They found that the measured $C(\ell)\sim\exp(-\ell/\xi)$, where the velocity correlation length ξ depends on the particle radius a and volume fraction ϕ_0 in a nontrivial power-law form $\xi\simeq a\phi_0^{-1/3}$. In this Rapid Communication we propose a set of coarse-grained equations of motion to describe concentration and velocity fluctuations in colloidal sedimentation. With these equations we find that colloidal sedimentation is analogous to high Rayleigh number, high Prandtl number turbulent convection [8,9]. Our model explains the experimental results by Segrè *et al.*, and also provides a coherent framework for the study of sedimentation dynamics in different colloidal systems.

To understand the basic principles governing the colloidal sedimentation, we consider a simple case of a dilute sedimenting suspension of hard spheres in a long tube of radius L. To separate the velocity fluctuation $\delta\mathbf{u}$ from the mean settling velocity $\overline{v}$, we choose a uniform suspension with $\overline{v}=0$ as our reference system. This can be achieved by using a fluidized bed, in which the mean settling velocity is opposed and canceled by a solvent velocity. It has been suggested [7,10] that velocity fluctuations in a sedimenting suspension may arise from fluctuations of the local particle concentration. Therefore, we model the colloidal sedimentation with a coarse-grained Navier-Stokes equation. The fluid velocity $\delta\mathbf{u}$ and pressure δp at a point $\mathbf{x}$ satisfy the creeping flow equation [3,6]

$$\nabla\,\delta p(\mathbf{x})-\eta\nabla^2\delta\mathbf{u}(\mathbf{x})=\mathbf{f}\delta n(\mathbf{x}), \tag{1}$$

where η is the viscosity of the fluid and δn $[=n(\mathbf{x})-\overline{n}]$ represents the fluctuation of the particle number density $n(\mathbf{x})$ about its mean $\overline{n}$. In the above, $\mathbf{f}=(4\pi/3)a^3\Delta\rho\mathbf{g}$ is the buoyancy force acting on a particle of radius a, where $\mathbf{g}$ is the gravitational acceleration and $\Delta\rho=\rho_p-\rho_s$ is the density difference between the particle and the solvent. In writing Eq. (1) we have assumed that the fluid volume element δV is a coarse-grained volume, which is large enough to contain many particles but is small enough such that the particle distribution inside δV is uniform. In this case, we have $\mathbf{f}\delta n(\mathbf{x})=\Delta\rho\mathbf{g}[\phi(\mathbf{x})-\phi_0]$, where $\phi(\mathbf{x})$ is the particle volume fraction and ϕ_0 is its mean value. Note that $\delta\mathbf{u}$ in Eq. (1) represents the velocity fluctuation of the solution, which includes both the particles and the solvent. In the coarse-grained sense, velocity fluctuations of the particles and the solvent are statistically the same as $\delta\mathbf{u}$. Because local concentration fluctuations are collective motions of the particles, Eq. (1) is different from usual equations of motion for individual particles.

Nondimensionalizing Eq. (1) with respect to the length L, the time L^2/D, and the concentration ϕ_0, we have

$$-\frac{1}{\sigma}\nabla\,\delta p(\mathbf{x})+\nabla^2\delta\mathbf{u}(\mathbf{x})=\mathrm{Ra}\;\phi(\mathbf{x})\hat{\mathbf{z}}, \tag{2}$$

where the unit vector $\hat{\mathbf{z}}$ is directed upward opposite to the direction of $\mathbf{g}$, and δp has included a term, $-\Delta\rho g \phi_0 z$, to absorb contributions from the constant forcing term $-\Delta\rho \mathbf{g}\phi_0$. In Eq. (2) the Rayleigh number is defined as

$$\mathrm{Ra} = \Delta\rho g \phi_0 L^3/(\eta D), \tag{3}$$

where D is an effective diffusion constant of the particles. The Schmidt number σ is given by $\sigma = \nu/D$ with ν being the kinematic viscosity of the fluid. For a dilute suspension of small colloidal particles, D is approximately equal to the particle self-diffusion constant $D_s = k_B T/(6\pi\eta a)$, where $k_B T$ is the thermal energy. For large non-Brownian particles, however, the effect of thermal agitations is negligible and their diffusionlike motion is produced by the hydrodynamic interactions between the particles [1]. Nicolai *et al.* have shown [5] that the hydrodynamic diffusivity has the form $D_h \simeq 5aU_0$, where $U_0 = 2a^2\Delta\rho g/(9\eta)$ is the Stokes velocity.

With the hydrodynamic diffusivity D_h, Eq. (3) becomes

$$\mathrm{Ra} = 0.9\phi_0 \left(\frac{L}{a}\right)^3. \tag{4}$$

It should be mentioned that while it is canceled out in Ra, $\Delta\rho$g is needed so that D_h can be used to describe the hydrodynamic diffusion of the settling particles at small length scales. Equation (2), together with the continuity equation for an incompressible fluid

$$\nabla \cdot \delta\mathbf{u} = 0 \tag{5}$$

and the advective mass diffusion equation

$$\partial_t \phi + (\delta\mathbf{u}\cdot\nabla)\phi = \nabla^2\phi, \tag{6}$$

complete the description of concentration and velocity fluctuations in colloidal sedimentation. Under the Boussinesq approximation [11], small density changes of the fluid due to concentration fluctuations have been neglected in Eq. (5). We notice that similar equations of motion have been used previously to describe several special problems in sedimentation [12,13].

It is evident that Eqs. (2)–(6) are the same as those for buoyancy-driven convection [9,11]. Velocity and concentration fluctuations in colloidal sedimentation are therefore analogous to those in buoyancy-driven convection, and they are completely controlled by the two dimensionless parameters Ra and σ, once the boundary conditions are specified. In a typical convection experiment, a constant temperature (or concentration) difference is usually maintained across a fluid layer of thickness L. It is this large-scale temperature gradient that drives the convective flow. A finite critical value Ra_c ($\simeq 2000$) is required for the onset of the convection instability with a vertical temperature gradient, but $\mathrm{Ra}_c \simeq 0$ for thermal convection in a vertical slot heated from the side [11]. In the latter case, the fluid is absolutely unstable to temperature or concentration perturbations.

In most sedimentation experiments, however, no large-scale concentration gradient is imposed upon the sample. Instead, a constant flux of particles (or solvent) is maintained throughout the bulk region. While no stability analysis is available at the moment for colloidal sedimentation, we believe that velocity and concentration fluctuations can be induced by an instability due to the mean settling flow. Regions having more particles become heavier than the average, and they can immediately induce velocity fluctuations in the direction parallel to gravity. Furthermore, Crowley [14] has shown that such a flow is unstable against concentration fluctuations at least for a special case of a one-dimensional array of falling spheres. Batchelor *et al.* [13] have studied (transient) ''homogeneous buoyancy-generated turbulence'' for colloidal suspensions with finite particle sizes. While Batchelor's theory describes the decay of fluctuations to states with average mean-square fluctuations of zero amplitude, the mean-square density fluctuations are not zero even in equilibrium and can serve as a source for continued homogeneous buoyancy-generated turbulence. The real mechanism for the generation of density fluctuations in sedimentation is not well understood, but is generally believed to be the spontaneous number fluctuations in the colloid suspension. An issue that remains to be resolved is that the convection instability usually takes place first at the largest scale of the system, whereas the spontaneous number fluctuations occur at all length scales.

Nonuniform particle concentrations can also result from shear-induced particle migration. Because of the hydrodynamic interactions between three or more particles, non-Brownian particles do not move along streamlines but instead exhibit diffusionlike motions as they tumble around each other. As a result, the particles in the high-shear region will migrate to low-shear regions, because they have a higher frequency of encounters with the surrounding high-shear particles [1]. Koh *et al.* [15] have shown that in a channel flow the particle concentration becomes higher in the central region of the channel and reaches a steady profile, so that the particle's hydrodynamic diffusion toward the center, due to the gradient of the shear rate, is balanced by the hydrodynamic diffusion away from the center, due to the gradient in particle concentration. For particles settling in a quiescent fluid, the downward volume flux of particles must be balanced by an upward volume flux of the solvent. In a long cylindrical tube, this back flow of the solvent must have a parabolic velocity profile when the particle density and resulting force density on the solvent is (initially) uniform. Thus, shear-induced particle migration is possible. Increasing particle densities in the tube center will favor downward convection in that region, so it is not at all clear how the system resolves these opposite tendencies. A detailed stability analysis of Eqs. (2)–(6), including hydrodynamic diffusion [in Eq. (6)], should be done. Experimentally, the sediment-supernatant interface is observed to be flat. If the sedimentation instability is induced by an unstable concentration profile $\phi(r)$, it becomes natural that Ra scales with ϕ_0 because $\phi(r)$ is proportional to ϕ_0. Clearly, this instability is fed by the potential energy of the settling particles. To verify the mechanism for the sedimentation instability, one needs to measure the concentration profile of a settling suspension of non-Brownian particles at low Ra.

In the discussion below, we will assume that such an instability exists in colloidal sedimentation at low Ra, and focus our attention on velocity and concentration fluctuations at high Ra far beyond Ra_c. Recent studies of Rayleigh-

Bénard convection have demonstrated [9] that high-Ra turbulent convection does not depend on details of the instability at low Ra. As will be shown below, turbulent convection is characterized by the emergence of new length scales in the velocity and concentration fields. We now estimate typical values of Ra and σ in colloidal sedimentation. In the experiment by Segrè *et al.* [7], the particle's radius $a\simeq 8\ \mu$m, Stokes velocity $U_0\simeq 6.5\ \mu$m/s, volume fraction $\phi_0\simeq 0.05$, and the characteristic sample size $L\simeq 1$ cm. With these experimental values we find $D_h\simeq 2.6\times 10^{-6}$ cm^2/s, Ra$\simeq 8.8\times 10^7$, and $\sigma\simeq 3800$. The Schmidt number σ is equivalent to the Prandtl number in thermal convection. Colloidal sedimentation is therefore associated with high Rayleigh number, high Prandtl number turbulent convection.

To understand the sedimentation dynamics, it is helpful to distinguish two characteristic length scales in convection: the viscous dissipation length δ_v and the diffusive dissipation length δ_d. The values of δ_v and δ_d are determined, respectively, by the transition Reynolds number $\mathrm{Re}_c=\widetilde{\delta u}\,\delta_v/\nu$ and the transition Péclet number $\mathrm{Pe}_c=\widetilde{\delta u}\,\delta_d/D_h$. Here $\widetilde{\delta u}$ is the rms value of the velocity fluctuation δu averaged over a volume of δ_v^3 (or δ_d^3). It is the ratios of these lengths to each other and to the sample size L that determine the flow state of the system [8]. For high-Ra, high-σ turbulent convection, one anticipates that the flow consists of three different regions: (i) $a<\ell<\delta_d$, (ii) $\delta_d<\ell<\delta_v$, and (iii) $\delta_d<\ell<L$. In region (i), molecular viscosity and hydrodynamic diffusivity determine the momentum and mass transport processes, respectively, and hence the particle distribution remains uniform without any large fluctuations. In region (ii), turbulent (or eddy) diffusivity and molecular viscosity are dominant, and thus large fluctuations in particle concentration are expected but the velocity field remains relatively smooth. Finally, in region (iii), turbulent diffusivity and viscosity both dominate over the corresponding hydrodynamic and molecular processes. In this case, one expects to see large fluctuations both in particle concentration and in velocity at different length scales. For most colloidal suspensions, however, their sedimentation velocity is so small that region (iii) may not exist (i.e., $\delta_d\geqslant L$). In this case, the bulk region of the fluid is dominated by the molecular viscosity and the local Reynolds number $\mathrm{Re}(\ell)=\widetilde{\delta u}\,\ell/\nu$ is smaller than Re_c everywhere. Note that $\mathrm{Re}(\ell)<\mathrm{Re}_c$ does not imply the absence of turbulence. When Ra is large, strong concentration fluctuations (and hence large buoyancy forces) can still drive the system into a state of chaotic motion.

Convective turbulence in region (ii) is different from that in region (iii), which can be realized in many low Prandtl number fluids, and has been intensively studied in recent years [9]. Many temperature [16] and velocity [17] measurements have been carried out in turbulent bulk regions and near viscous and thermal boundary layers. In contrast to the great number of low-σ experiments, experimental information about high-σ turbulent convection is limited. Many years ago, Kraichnan [8] proposed a mixing-length theory for turbulent convection at arbitrary Prandtl numbers. For high-σ turbulent convection in region (ii), he obtained scaling relations for the temperature variance $\widetilde{\delta T}$ (or, equivalently, the concentration variance $\widetilde{\delta\phi}$), the velocity variance $\widetilde{\delta u}$, and the diffusive dissipation length δ_d. In the following we use Kraichnan's theory to calculate small-scale properties of concentration and velocity fluctuations in colloidal sedimentation.

We first discuss the length δ_d, above which velocities become large and concentration fluctuations are transported by convection. This occurs when the local Péclet number $\mathrm{Pe}=\widetilde{\delta u}\,\ell/D_h$ becomes larger than Pe_c. Recent thermal convection experiments have shown [18] that while turbulent mixing creates, on average, an isothermal fluid in the turbulent bulk region, large temperature fluctuations still remain in the region and the characteristic length scale associated with these fluctuations is of the order of δ_d. Therefore, the velocity correlation length ξ is determined by δ_d in region (ii). According to Kraichnan's theory [8], we have

$$\delta_d\simeq(2\pi^2\ \mathrm{Pe}_c^2)^{1/3}L\ \mathrm{Ra}^{-1/3}, \tag{7}$$

where the power law amplitude is expressed in terms of the numerical value of Pe_c. Priestley first explained why δ_d scales with $\mathrm{Ra}^{1/3}$ [19]. He argued that when Ra is large enough, δ_d should be a new length scale independent of L. With Eqs. (7) and (4), we immediately have $\xi\sim\delta_d\simeq L\ \mathrm{Ra}^{-1/3}\simeq a\phi_0^{-1/3}$. The mapping of colloidal sedimentation to turbulent convection, therefore, explains the experimental finding that $\xi\simeq 11a\phi_0^{-1/3}$ [20]. It also provides a physical interpretation for the existence of a velocity cutoff length, which prevents hydrodynamic dispersion coefficients from being divergent.

We now discuss the velocity variance $\widetilde{\delta u}$ in colloidal sedimentation. According to Kraichnan's theory [8],

$$\widetilde{\delta u}\simeq\frac{\mathrm{Pe}_c D_h}{\delta_d}\simeq\frac{\mathrm{Pe}_c D_h}{(2\pi^2\ \mathrm{Pe}_c^2)^{1/3}L\ \mathrm{Ra}^{-1/3}}. \tag{8}$$

Equation (8) states that at the transition Péclet number Pe_c, the mass flux due to hydrodynamic diffusion, $D_h\widetilde{\delta\phi}/\delta_d$, is approximately equal to that by convection, $\widetilde{\delta u}\,\widetilde{\delta\phi}$. Because $\delta_d\simeq a\phi_0^{-1/3}$ and $D_h\simeq aU_0$, we find from Eq. (8) that $\widetilde{\delta v}\sim\widetilde{\delta u}\simeq D_h/\delta_d\simeq U_0\phi_0^{1/3}$, which is independent of the sample size L. This result agrees well with the experimental finding that $\widetilde{\delta v}\simeq 2U_0\phi_0^{1/3}$ [7]. Another important quantity in colloidal sedimentation is the concentration variance $\widetilde{\delta\phi}$. Kraichnan's theory [8] predicts that the typical value of $\widetilde{\delta\phi}$ is of the order of ϕ_0. Because the sedimentation velocity is determined by the particle concentration, $\widetilde{\delta\phi}\simeq\phi_0$ implies $\widetilde{\delta v}\simeq\bar{v}$. This explains another important experimental observation that velocity fluctuations in colloidal sedimentation are of the same order as the mean settling velocity [5,21].

As shown in Eqs. (7) and (8), Kraichnan's theory not only predicts definite values of the scaling exponents, but also provides a rough estimate for the amplitude of the power laws. These amplitudes are expressed in terms of the numerical value of Pe_c, which is independent of any dynamic variables in the problem. Indeed, from the definition of Pe_c and the measured values of D_h, ξ, and $\widetilde{\delta v}$, we find $\mathrm{Pe}_c\simeq\widetilde{\delta v}\,\xi/D_h\simeq 4.4$, which is a constant independent of ϕ_0 and a. Recently, Goldstein and Chiang [22] have carried out a convection experiment with $\sigma\simeq 2750$. From their transport data one finds $\mathrm{Pe}_c \simeq 4.7$, which is in good agreement with the sedimentation value. Using Eqs. (7) and (8), together with the measured value of $\mathrm{Pe}_c \simeq 4.4$, we obtain $\delta_d \simeq 7.5 a \phi_0^{-1/3}$ and $\widetilde{\delta u} \simeq 2.9 U_0 \phi_0^{1/3}$. This calculation demonstrates that our model is even capable of predicting correct values of the power-law amplitudes. In the above calculation, we used the measured values of $\widetilde{\delta v}$, ξ, and D_h in the direction parallel to gravity. We notice that the measured hydrodynamic diffusivity, D_h, changes with the direction relative to gravity [5], whereas the convective diffusivity, $\widetilde{\delta v} \xi$, is less sensitive to the direction of sedimentation [7]. The measured D_h is anisotropic because it couples to the mean sedimentation velocity U_0. Turbulent convection, on the other hand, is expected to be isotropic because there is no preferred direction in the large scale motion.

We thank P. M. Chaikin for useful discussions. This work was supported by the National Aeronautics and Space Administration under Grant No. NAG3-1852. In addition, P. Tong was also supported in part by the National Science Foundation under Grant No. DMR-9623612.

[1] R. H Davis, J. Fluid Mech. **310**, 325 (1996), and references therein.

[2] W. B. Russel, D. A. Saville, and W. R. Schowalter, *Colloidal Dispersions* (Cambridge University Press, Cambridge, England, 1989).

[3] R. E. Caflisch and J. H. C. Luke, Phys. Fluids **28**, 759 (1985).

[4] A. J. C. Ladd, Phys. Rev. Lett. **76**, 1392 (1996).

[5] H. Nicolai and E. Guazzelli, Phys. Fluids **7**, 3 (1995); H. Nicolai *et al.*, *ibid.* **7**, 12 (1995).

[6] D. L. Koch and E. S. G. Shaqfeh, J. Fluid Mech. **224**, 275 (1991).

[7] P. N. Segrè, E. Herbolzheimer, and P. M. Chaikin, Phys. Rev. Lett. **79**, 2574 (1997).

[8] R. H. Kraichnan, Phys. Fluids **5**, 1374 (1962).

[9] E. D. Siggia, Annu. Rev. Fluid Mech. **26**, 137 (1994).

[10] E. J. Hinch, in *Disorder and Mixing*, edited by E. Guyon *et al.* (Kluwer, Dordrecht, 1988), p. 153.

[11] S. Chandrasekhar, *Hydrodynamic and Hydromagnetic Stability* (Dover, New York, 1981).

[12] A. E. Hosoi and T. Depont, J. Fluid Mech. **328**, 297 (1996); S. E. Esipov, Phys. Rev. E **52**, 3711 (1995); W. D. Hill, R. R. Rothfus, and Kun Li, Int. J. Multiphase Flow **3**, 561 (1977).

[13] G. K. Batchelor, V. M. Canuto, and J. R. Chasnov, J. Fluid Mech. **235**, 349 (1992).

[14] J. M. Crowley, J. Fluid Mech. **45**, 151 (1971).

[15] C. J. Koh, P. Hookham, and L. G. Leal, J. Fluid Mech. **266**, 1 (1994).

[16] B. Castaing *et al.*, J. Fluid Mech. **204**, 1 (1989); T. H. Solomon and J. P. Gollub, Phys. Rev. Lett. **64**, 2382 (1990); A. Belmonte, A. Tilgner, and A. Libchaber, Phys. Rev. E **50**, 269 (1994).

[17] Y. Shen, K.-Q. Xia, and P. Tong, Phys. Rev. Lett. **75**, 437 (1995); K.-Q. Xia, Y.-B. Xin, and P. Tong, *ibid.* **77**, 1266 (1996).

[18] G. Zocchi, E. Moses, and A. Libchaber, Physica A **166**, 387 (1990).

[19] C. H. B. Priestley, *Turbulent Transfer in the Low Atmosphere* (University of Chicago Press, Chicago, 1959).

[20] In a more recent measurement, Segrè *et al.* changed the particle radius a from 7.8 μm to 3 μm and verified that ξ indeed scales as $a\phi_0^{-1/3}$.

[21] J. M. Ham and G. M. Homsy, Int. J. Multiphase Flow **14**, 533 (1988); J.-Z. Xue *et al.*, Phys. Rev. Lett. **69**, 1715 (1992).

[22] R. J. Goldstein and H. D. Chiang, J. Fluid Mech. **213**, 111 (1990).