RAPID COMMUNICATIONS

PHYSICAL REVIEW E VOLUME 51, NUMBER 5 MAY 1995

Statistical theory of sedimentation of disordered suspensions

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(Received 25 July 1994)

An analytical treatment for the sedimentation rate of disordered suspensions is presented in the context of a resistance problem. From the calculation it is confirmed that the lubrication effect is important in contrast to the suggestion by Brady and Durlofsky [J. F. Brady and L. J. Durlofsky, Phys. Fluids **31**, 717 (1988)]. The calculated sedimentation rate agrees well with the experimental results throughout the range of the volume fraction.

PACS number(s): 05.60.+w, 05.40.+j, 47.55.Kf, 83.70.Hq

The sedimentation of disordered suspensions is important in both technology and the laboratory [1,2]. The role of sedimentation is relevant to current topics of statistical mechanics such as fluidized beds of gas-solid or liquid-solid mixtures [3–5], and density waves in granular flows in vertical tubes [6]. We believe the subject is a fundamental one in fluid mechanics [7]. The rate of sedimentation for disordered suspensions under gravity has yet to be determined theoretically except for a problem for dilute spheres with hard core interactions at a small Reynolds number [1,8].

Our present understanding of theoretical studies of sedimentation of monodisperse random suspensions can be summarized as follows. Batchelor [8] has calculated the sedimentation rate in the dilute limit of hard core particles with the radius a based on the following assumptions: (i) The rate can be obtained from the combination of the mobility matrix of two particles and the two-body correlation function $g_{eq}(r)$ where r is the relative distance of particles, and (ii) the correlation function is assumed to be $g_{eq}(r) = \theta(r-2a)$, where $\theta(x)$ is the step function $\theta(x)=1$ for $x \geqslant 0$ and $\theta(x)=0$ otherwise. His result at the volume fraction ϕ can be written as $U(\phi)/U_0 = 1-6.55\phi + O(\phi^2)$ for $\phi \rightarrow 0$, where $U(\phi)$ is the sedimentation velocity at ϕ and U_0 is the equilibrium sedimentation velocity of one particle. The result of Batchelor consists of two parts: one is $1-5\phi$ from the Rotne-Prager tensor which represents the effects of long-range hydrodynamic interaction, and another is -1.55ϕ from the lubrication, the hydrodynamic repulsive force. Extensions of this dilute theory to concentrated suspensions require an account of many-body hydrodynamic interactions. A generalization [9], based on the method of O'Brien [10], predicts negative sedimentation rate for $\phi > 0.27$. Brady and Durlofsky [11] have also obtained a negative sedimentation rate for $\phi > 0.23$ when they adopt the well accepted correlation function $g_{eq}(r)$ for concentrated suspensions. As a result, they claim that the Rotne-Prager approximation actually captures the correct features of sedimentation and ignore all of the contributions from the lubrication force. We feel, however, the statement by Brady and Durlofsky [11] is unacceptable, because there is no reason to ignore lubrication effects in the dilute limit [8]. On the other hand, Beenakker and Mazur [12,13] also calculated the sedimentation rate based on an effective medium approximation and multipole expansions. Although they did not present an explicit expression of the sedimentation rate, Ladd [14] indicated that their result is better than the result by Brady and Durlofsky [11] for concentrated suspensions. In this Rapid Communication, we wish to demonstrate the relevance of the lubrication force and improve the theory by Brady and Durlofsky [11]. We also clarify the relationship between our theory and that by Beenakker and Mazur [11,12].

The problem of sedimentation of N particles with radius a at low Reynolds numbers is equivalent to obtaining the resistance matrix R or the mobility matrix M in

$$\mathbf{U} = \frac{1}{6\pi\mu a}\mathsf{M}\cdot\mathbf{F}, \quad \mathsf{M} = \mathsf{R}^{-1}, \tag{1}$$

where $\mathbf{U}$ and $\mathbf{F}$ denote the sets of the velocity field of N particles and the force exerted on N particles, respectively, and μ is the shear viscosity. These mobility and resistance problems are not easy to solve even numerically. One of the most successful numerical methods, the Stokesian dynamics, has been developed by Brady and co-workers [15,16]. The extension by Ladd [14] also follows an algorithm similar to the Stokesian dynamics. They decouple the resistance matrix into the far-field part $(\mathsf{M}^{\infty})^{-1}$ and the lubrication part R^{lub} as

$$\mathsf{R} = (\mathsf{M}^{\infty})^{-1} + \mathsf{R}^{lub}, \tag{2}$$

where R^{lub} is calculated by the pairwise additive expression of the two-body lubrication matrix $\mathsf{R}_{2B}^{lub} = \mathsf{R}_{2B} - (\mathsf{M}_{2B}^{\infty})^{-1}$. The resistance matrix is calculated as a function of the particle configuration at each numerical step. Then the force exerted on spheres and consequently the equation of motion are obtained. The success in the Stokesian dynamics suggests that the problem for sedimentations should be considered based on a resistance picture. In fact, some unphysical results of simulations based on a mobility picture support this statement. We may understand the relevance of a resistance picture as follows. Since the contribution of the lubrication is proportional to the number of particles, as will be shown, the

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direct addition of the lubrication for the mobility cannot avoid a negative sedimentation rate. In other words, the linear contribution of the lubrication to the drag is reasonable, while the linear addition of the lubrication to the mobility cannot produce any nonlinear complicated motion of particles in experiments.

Thus we are not surprised by the failure of direct generalizations of Batchelor's theory which is described as a mobility problem. We must calculate the sedimentation rate in the context of a resistance problem. The problem is thus reduced to obtaining $\langle(\mathsf{M}^{\infty})^{-1}\rangle+\langle\mathsf{R}^{lub}\rangle$, where the angular brackets represent the average over the particle configurations. Note that $\langle(\mathsf{M}^{\infty})^{-1}\rangle$ and $\langle\mathsf{R}^{lub}\rangle$ are the scalar quantities. The far-field part can be calculated from $\langle(\mathsf{M}^{\infty})^{-1}\rangle\simeq\langle\mathsf{M}^{\infty}\rangle^{-1}=\tilde{M}(k=0)^{-1}$, where $\tilde{M}(k)$ is defined by

$$\tilde{M}(k)=1+n\int_V e^{i\mathbf{k}\cdot\mathbf{r}_{12}}[g_{eq}(\mathbf{r}_{12})-1]\hat{\mathbf{k}}\cdot\mathsf{G}(\mathbf{r}_{12})\cdot\hat{\mathbf{k}}d\mathbf{r}_{12}. \quad (3)$$

Here $\hat{\mathbf{k}}=\mathbf{k}/k$, $\mathbf{r}_{12}$ is the relative position of the particles 1 and 2, and n is the number density of particles. The explicit representation of the Fourier component of the tensor $\mathsf{G}=\{G_{ij}\}$ is given by [13,17]

$$G_{ij}(\mathbf{k})=6\pi a\,\frac{j_0(ka)^2}{k^2}\left(\delta_{ij}-\frac{k_ik_j}{k^2}\right) \quad (4)$$

with the spherical Bessel function $j_0(ka)$. For later discussion we drop the subscripts to $\mathbf{r}_{12}$ and assume the isotropy of systems as $g_{eq}(r=|\mathbf{r}|)$.

The correlation function $g_{eq}(r)$ can be approximated [16,17] by the equilibrium distribution function for hard sphere systems based on the Percus-Yevick approximation [18]. The Fourier transform of $g_{eq}(r)-1$, $h(k)$ is represented by [19]

$$h(k)=-\frac{4\pi a^3\tilde{c}(ka)}{1+3\phi\tilde{c}(ka)}, \quad (5)$$

where $\tilde{c}(x)$ is the direct correlation function which also depends on ϕ. The correlation function in (5) reduces to $g_{eq}(r)=\theta(r-2a)$ in the dilute limit. From (5) we can evaluate $\langle\mathsf{M}^{\infty}\rangle=(2/\pi)\int_0^{\infty}dx[(\sin x)/x]^2[1+3\phi\tilde{c}(x)]^{-1}$ numerically. Brady and Durlofsky [11] evaluated this [20] by using the Laplace transform of the Percus-Yevick distribution function [21] and the method of O'Brien [10] as

$$\langle\mathsf{M}^{\infty}\rangle\simeq\frac{(1-\phi)^3}{(1+2\phi)}, \quad (6)$$

which is a correct evaluation of the contribution from the far-field part.

Now, we evaluate the contribution from $\langle\mathsf{R}^{lub}\rangle$. For simplicity of the argument, we neglect contributions from higher order moments such as torque and shear. Since $\langle\mathsf{R}^{lub}\rangle$ is evaluated from a pairwise additive approximation, $\langle\mathsf{R}^{lub}\rangle$ is represented by

$$\langle\mathsf{R}^{lub}\rangle=n\int_V d\mathbf{r}g_{eq}(r)\hat{\mathbf{k}}\cdot[\mathsf{A}_{11}+\mathsf{A}_{12}-\{(\mathsf{M}_{2B}^{\infty})^{-1}{}_{11}+(\mathsf{M}_{2B}^{\infty})^{-1}{}_{12}\}]\cdot\hat{\mathbf{k}}. \quad (7)$$

The tensor $\mathsf{A}_{\alpha\beta}$ is a part of R_{2B} and its subscripts represent the particles. The tensor $\mathsf{A}_{11}+\mathsf{A}_{12}$, thus, is given by

$$\mathsf{A}_{11}+\mathsf{A}_{12}=\begin{pmatrix} Y_{11}+Y_{12} & 0 & 0\\ 0 & Y_{11}+Y_{12} & 0\\ 0 & 0 & X_{11}+X_{12}\end{pmatrix}, \quad (8)$$

where the explicit representations of X_{ij} and Y_{ij} are given by Jeffrey and Onishi [22] as a series expression. On the other hand, $(\mathsf{M}_{2B}^{\infty})_{11}$ is the unit tensor and $(\mathsf{M}_{2B}^{\infty})_{12}$ is the Rotne-Prager tensor which is represented by

$$(\mathsf{M}_{2B}^{\infty})_{12}=x^{\infty}(r)\hat{\mathbf{r}}\hat{\mathbf{r}}+y^{\infty}(r)(\mathsf{I}-\hat{\mathbf{r}}\hat{\mathbf{r}}), \quad (9)$$

where $x^{\infty}(r)=\frac{3}{2}(r/a)^{-1}-(r/a)^{-3}$ and $y^{\infty}(r)=\frac{3}{4}(r/a)^{-1}+\frac{1}{2}(r/a)^{-3}$. The tensor $(\mathsf{M}_{2B}^{\infty})^{-1}{}_{11}+(\mathsf{M}_{2B}^{\infty})^{-1}{}_{12}$ can be readily calculated as

$$(\mathsf{M}_{2B}^{\infty})^{-1}{}_{11}+(\mathsf{M}_{2B}^{\infty})^{-1}{}_{12}=X^{\infty}(r)\hat{\mathbf{r}}\hat{\mathbf{r}}+Y^{\infty}(r)(\mathsf{I}-\hat{\mathbf{r}}\hat{\mathbf{r}}), \quad (10)$$

where $X^{\infty}(r)=[1+x^{\infty}(r)]^{-1}$ and $Y^{\infty}(r)=[1+y^{\infty}(r)]^{-1}$. Thus the average of the contribution from the lubrication part is described by

$$\langle\mathsf{R}^{lub}\rangle=\phi\int_2^{\infty}dzz^2g_{eq}(r)W(z)\ , \quad (11)$$

where $z=r/a$ and

$$W(z)=X_{11}+X_{12}+2Y_{11}+2Y_{12}-\frac{6z^3(-2+5z^2+4z^3)}{(-2+3z^2+2z^3)(2+3z^2+4z^3)}. \quad (12)$$

With the aid of the exact result by Jeffrey and Onishi [22] $W(z)$ can be evaluated as

$$W(z)=\frac{21}{4}\frac{1}{z^4}-\frac{789}{64}\frac{1}{z^5}+O\left(\frac{1}{z^6}\right) \quad (13)$$

for $z\gg1$. Thus we can evaluate $\langle\mathsf{R}^{lub}\rangle$ by the numerical integral. For practical purposes, it is convenient to have an explicit expression for $\langle\mathsf{R}^{lub}\rangle$. If we assume $g_{eq}(r)=\theta(r-2a)$, $\langle\mathsf{R}^{lub}\rangle$ is approximately represented by

$$\langle\mathsf{R}^{lub}\rangle\simeq\phi\int_2^{20}dzz^2W(z)+\phi\int_{20}^{\infty}dzz^2\left(\frac{21}{4}\frac{1}{z^4}-\frac{789}{64}\frac{1}{z^5}\right)\simeq1.492\phi. \quad (14)$$

When we compare the result (14) with the one obtained with the aid of the Percus-Yevick distribution function for $g_{eq}(r)$, we find that the two results have no significant difference (see Fig. 1). This statement is applicable to the calculation for the lubrication part of the mobility matrix as $\langle\mathsf{M}^{lub}\rangle\simeq-1.55\phi$. We thus confirm that the contribution from the lubrication is insensitive to the form of $g_{eq}(r)$ and

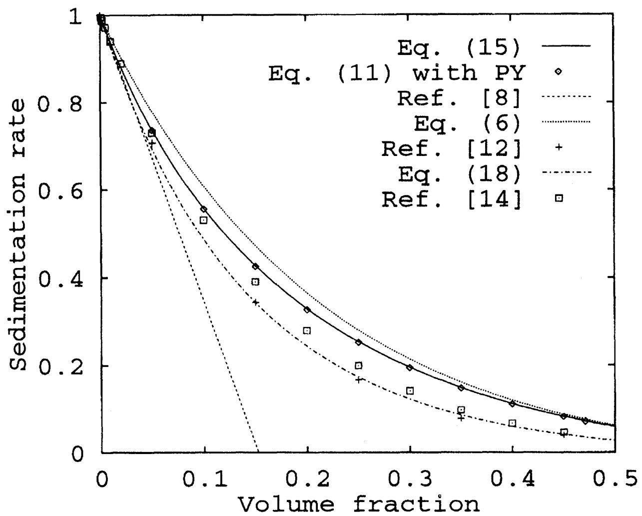

FIG. 1. The comparison of several theoretical and numerical predictions of the sedimentation rate $U(\phi)/U_0$ as functions of ϕ. For Eq. (11) with PY, we use the Percus-Yevick distribution function for $g_{eq}(r)$ in (11) to evaluate $\langle \mathsf{R}^{lub}\rangle$. The result of Ref. [14] is obtained from his precise simulation. Reference [12] is from their Fig. 2 with $k=0$ and its approximate expression is given by (18).

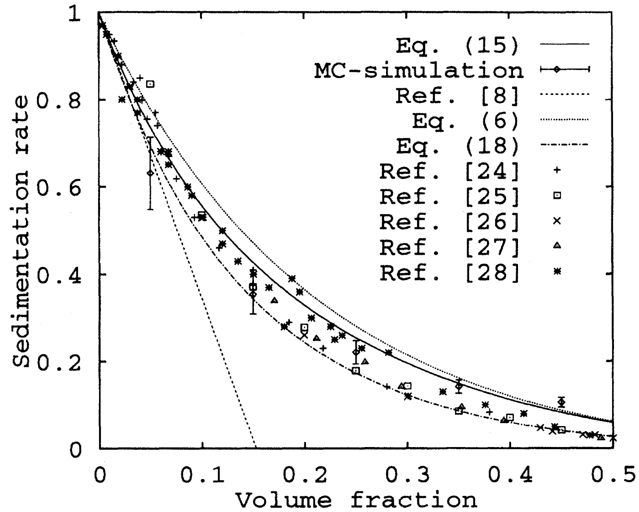

FIG. 2. The comparison of several theoretical results with experimental results for $U(\phi)/U_0$. We also plot the data of our Monte Carlo simulation. See the text for the details.

is proportional to ϕ. Thus we should solve the problem in the context of a resistance problem to avoid a negative sedimentation rate.

From (6) and (14) we obtain

$$\frac{U}{U_0}\simeq\frac{1}{\langle \mathsf{M}^{\infty}\rangle^{-1}+\langle \mathsf{R}^{lub}\rangle}=\frac{(1-\phi)^3}{1+2\phi+1.492\phi(1-\phi)^3}. \quad (15)$$

As will be shown, this result is sufficiently close to experimental values. The dilute limit of our result $U/U_0=1-6.49\phi+O(\phi^2)$ is slightly different from Batchelor's result $U/U_0=1-6.55\phi+O(\phi^2)$. This discrepancy comes from the relation $\mathsf{R}^{lub}\neq(\mathsf{M}^{lub})^{-1}$. The true dilute limit should be calculated under the considerations of all of higher order moments [23]. It is worthwhile, however, to indicate that our theory essentially resolves the contradiction about contributions from the lubrication in the result by Brady and Durlofsky [11].

Now we compare our result with that by Beenakker and Mazur [12]. They rewrite the renormalized (4) as

$$\tilde{M}_{\gamma_0}(k)=1+\hat{\mathbf{k}}\cdot \mathsf{G}_{\gamma_0}(\mathbf{r}=0)\cdot\hat{\mathbf{k}} +n\int d\mathbf{r}e^{i\mathbf{k}\cdot\mathbf{r}}\hat{\mathbf{k}}\cdot \mathsf{G}_{\gamma_0}(\mathbf{r})\cdot\hat{\mathbf{k}}\{g_{eq}(r)-1\}, \quad (16)$$

where $\mathsf{G}_{\gamma_0}(\mathbf{r})$ is given by

$$\mathsf{G}_{\gamma_0}(r)=\tilde{\mathsf{G}}(r)-\int\frac{d\mathbf{k}}{(2\pi)^3}e^{i\mathbf{k}\cdot\mathbf{r}}\frac{\phi S_{\gamma_0}(ka)}{1+\phi S_{\gamma_0}(ka)}\mathsf{G}(k). \quad (17)$$

Here $S_{\gamma_0}(x)$ is the structure factor and $\tilde{\mathsf{G}}(r)=0$ for $r=0$ and $\tilde{\mathsf{G}}(r)=\mathsf{G}(r)$ for $r\neq 0$. Substituting (5) into (16) and noting $\langle \mathsf{M}\rangle=\tilde{M}_{\gamma_0}(k=0)$, we obtain $\langle \mathsf{M}\rangle=(2/\pi)\int_0^\infty dx[(\sin x)/x]^2\{[1+\phi S_{\gamma_0}(x)][1+3\phi\tilde{c}(x)]\}^{-1}$. The function $S_{\gamma_0}(x)$ tends to $\frac{5}{2}$ for the dilute case and small x. In the dilute limit, the result by Beenakker and Mazur [12] is reduced to $U/U_0\simeq 1-\frac{15}{2}\phi+O(\phi^2)$, which is considerably far from Batchelor's result [8]. Even in concentrated cases, $S_{\gamma_0}(x)$ may still be replaced by $\frac{5}{2}$, although its actual expression is complicated. With the aid of (6) an approximate expression of Beenakker and Mazur [12] is given by

$$\frac{U}{U_0}\simeq\frac{(1-\phi)^3}{(1+2\phi)(1+5\phi/2)}. \quad (18)$$

From (18), it is easy to understand that Beenakker and Mazur [12] renormalize the Rotne-Prager tensor by taking into account the contribution from the structure factor. The deviation from Batchelor's result in the dilute limit suggests that they miss the quantitative description for the short-range force, because their effective field approximation includes only parts of the lubrication by a collection of ladder diagrams. Their theory, however, may be good for dense suspensions where the requirement for their approximation may be satisfied.

Let us compare theoretical results with experimental ones [24–28] (Fig. 2). We recognize that our theory improves the result by Brady and Durlofsky [11] and achieves good agreement with experiments. Therefore we conclude that the contribution from the lubrication force is small but relevant. For $\phi<0.2$, it seems that our result is better than that by Beenakker and Mazur [12]. In high concentration regions, however, our sedimentation rate is a little larger than the experimental values, while the prediction by Beenakker and Mazur [12] works well. This disagreement between our theory and experiments in concentrated regions seems to come from the neglect of higher order moments. A high sedimentation rate without higher order moments for a regular configuration of particles has been reported [15]. To check this tendency for random particle configurations we have performed a simulation for 50 particles based on the Stokesian dynamics, where we neglect the contributions from higher order multipole expansions. In our simulation the particle configuration is at random and we use an average of 100 configurations for each ϕ to calculate the sedimentation rate. When we neglect the statistical error, the tendency of high sedimentation rates in large ϕ coincides with that of our theory.

In conclusion, we have confirmed that the calculation of sedimentation rate should be performed in the context of a resistance problem. It is not surprising that the direct generalization of Batchelor's theory based on the mobility picture gives us wrong answers. Thus we should include the lubrication effects in contrast to the claim by Brady and Durlofsky [11]. Our method of including the lubrication force is an adequate systematic approach to extend the dilute theory. We demonstrate that the lowest order contribution to the sedimentation rate of the lubrication force becomes closer to experimental values than the Rotne-Prager approximation. The discrepancy between our calculation and experiments at high ϕ should be improved if we include the contribution from torque and other moments. The consecutive improvement of our calculation of the sedimentation rate will be reported elsewhere.

We thank T. Ohta and S. Sasa for stimulating discussion and Y. Oono for his critical reading and his useful comments. This work is, in part, supported by Foundation for Promotion of Industrial Science and by National Science Foundation Grant No. NSF-DMR-93-14938.

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