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The effect of weak Brownian rotations on particles in shear flow

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Axisymmetric particles in zero Reynolds number shear flow execute closed orbits. In this paper we consider the role of small Brownian couples in establishing a steady-state probability distribution for a particle being on any particular orbit. After presenting the basic equations, we derive an expression for the equilibrium distribution. This result is then used to calculate some bulk properties for a suspension of such particles, and these predicted properties are compared with available experimental observation.

1. Introduction

The motion of a single, small particle suspended in a Newtonian fluid which is undergoing a simple shear flow has been the subject of a considerable number of theoretical and experimental investigations which have spanned approximately fifty years. Interest in this problem stems mainly from its central role in the determination of the bulk properties of a dilute suspension composed of a large number of such particles in an ambient fluid. In a simple imposed shearing motion of the ambient fluid, the increased rate of dissipation which occurs due to the presence of a non-spherical particle is highly dependent upon its orientation, so that the rate of working for a given bulk motion of a dilute suspension will depend on the probability distribution of orientations among all the suspension particles.

The starting point for most of the recent work on the problem has been Jeffery's (1922) solution for the Stokes motion of a small rigid spheroid in a uniform shear flow of a Newtonian fluid. Jeffery showed that, in the absence of particle body forces or couples, a spheroid will translate with the velocity of the undisturbed fluid at the position of its centre, while its axis of revolution rotates in one of an infinite one-parameter family of possible periodic orbits. Hence, in order to determine the distribution of orientations among the particles of a dilute suspension we must first calculate the time-average distribution of orientations for each particular orbit, and second specify the statistical distribution of orbits amongst the particles. The first computation can be accomplished in a straightforward manner by employing the appropriate orbit equations (Anczurowski

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& Mason 1967) and hence, at least in principle, presents no theoretical difficulties. The distribution of orbits is more difficult to ascertain. According to Jeffery's analysis, a particle will remain indefinitely in any given orbit. Hence one might conclude that the distribution of particle orbits is completely determined by the initial distribution of orientations in the suspension. Of course, Jeffery's solution completely neglects such effects as fluid and particle inertia, Brownian motion and particle-particle interactions, all of which will be present in a suspension. Hence, an alternative view might be that these (presumably) small departures from undisturbed creeping flow will lead to a slow change in the orbit of a particle so that eventually a steady-state distribution of orbits may be established which is independent of the initial orientations of the particles.

If one adopts the former point of view, then the most plausible equilibrium distribution of orbits would seem to be that proposed by Eisenschitz (1932) based on the supposition that the particles are initially oriented over all possible directions with equal probability. Eisenschitz's distribution of orbits leads to a number of interesting results, among which is an effective viscosity (for steady shear flow) which is periodic in time. However, to the best of our knowledge, no experimental evidence either from direct measurement of particle orientations or from measurements of such bulk properties as the suspension viscosity has ever been reported in support of the Eisenschitz hypothesis. On the contrary, a number of investigators (Taylor 1923, Binder 1939, Mason & Manley 1956, Anczurowski & Mason 1967) have observed a slow drift through orbits for particles in a suspension, and one suspects that, given sufficient time, even isolated particles would show variations in orbit due to the influence of relevant effects which are neglected in Jeffery's analysis. Inertial effects, Brownian rotations, particle-particle interactions and non-Newtonian properties of the suspending fluid have all been proposed, at one time or another, as the primary cause of the observed drift through orbits for particles in suspension. Indeed, we believe that under appropriate conditions (i.e. particle size, particle concentration, shear rate, etc.) each of these effects could be of primary importance.

Jeffery himself suggested that a proper account of inertial effects would show that the particles ultimately assume the particular orientation corresponding to the minimum rate of energy dissipation. Taylor's (1923) early experiments seem to verify the minimum dissipation hypothesis for the case of spheroids. Subsequently, however, Saffman (1956) suggested that Taylor's results might have been due to some effect other than inertia, such as possible non-Newtonian character of the suspending fluid. Recently, Harper & Chang (1968) have reconsidered Jeffery's hypothesis for the case of small dumb-bell shaped particles, and have shown that the inertial drift across orbits eventually causes these particles to adopt an orientation corresponding to the *maximum* rate of energy dissipation. Hence it is clear that, at best, the minimum dissipation hypothesis has limited applicability. In any event, for many cases of interest, the particle Reynolds number is not sufficiently large for the influence of inertia on the particle orbit to dominate the effects of Brownian motion, particle-particle interactions or non-Newtonian properties of the suspending fluid.

Recently, Mason and coworkers (Manley & Mason 1956, Anczurowski &

Mason 1967) have measured the equilibrium distributions of orbits for suspensions of rods and disks with volume concentrations of order 10^{-4} and particle aspect ratios ranging from 0·26 to 20·8. Invariably, the measured distribution differed significantly from that predicted by either Eisenschitz or Jeffery, in fact lying somewhere between these two predictions. In addition, an apparent limiting distribution of orbits was obtained corresponding to very small volume concentrations of suspended particles. It was suggested, on the basis of observations of individual particles within the suspension, that particle-particle interactions were primarily responsible for the shift in the distribution of orbits from that corresponding to the Eisenschitz hypothesis. By inference, it could seemingly be concluded that the particle-particle interactions continue to be dominant even for vanishingly small concentrations of suspended particles.

Nevertheless, when the particles are sufficiently small, random rotations by Brownian motion clearly play a role in determining the orientation of the particles, and one would expect, intuitively, that this effect ultimately becomes more important than particle-particle interactions as the suspension is made more dilute. This intuitive motion might seem to be in substantial disagreement with two of Mason's experimental observations; first that an orbit distribution is found which appears to be asymptotically valid for small concentrations but is nevertheless apparently determined by particle-particle interactions, and second that single suspended particles appear to rotate with fixed orbits. With regard to the first of these points, it is unfortunately not entirely clear how to estimate the relative importance of the two competing disturbance mechanisms, since the details of the interaction problem have not yet been satisfactorily resolved. Nevertheless, as will be evident later, it seems likely that for the relatively large particles employed by Mason, the volume concentration was never sufficiently small for the Brownian motion effect to become noticeable. As to the constant orbit result for single particles, we note that when a particle is sufficiently large to be readily observable, the time scale for the drift across orbits due to Brownian motion would be exceedingly large and hence possibly not detectable even in experiments of long duration.

The case of strong Brownian motion acting on axisymmetric particles has been treated in detail by Burgers (1938). In this paper we derive an expression for the equilibrium distribution of orbit constants when the effect of Brownian motion is everywhere suitably small, but nevertheless dominant over inertial and particle-particle interaction effects.

2. The basic equations

We begin by considering a single spheroidal particle suspended in an ambient fluid which is undergoing a uniform shearing motion defined by

$$u = \gamma y; \quad v = w = 0. \tag{1}$$

The origin of the x, y, z co-ordinate axis system is assumed to be fixed at the centre of the particle. The orientation of the particle axis of revolution is described by the polar angles θ_1 and ϕ_1 shown in figure 1. Jeffery has shown that if inertial and Brownian motion affects are completely neglected, then the motion of the axis of revolution of the particle is described (apart from a simple translation which we have neglected by defining our shear flow relative to axes fixed at the centre of the particle) by

$$\dot{\phi}_1 = \frac{\gamma}{r^2+1}(r^2\cos^2\phi_1 + \sin^2\phi_1), \tag{2a}$$

$$\dot{\theta}_1 = \frac{\gamma(r^2-1)}{4(r^2+1)}\sin 2\phi_1 \sin 2\theta_1. \tag{2b}$$

Here r is the axis ratio of the particles, a/b, where a and b are the semi-diameters measured parallel and perpendicular, respectively, to the axis of revolution. Bretherton (1962) has shown that, with the exception of certain very long particles, the creeping motion of any rigid body of revolution in a simple shear flow

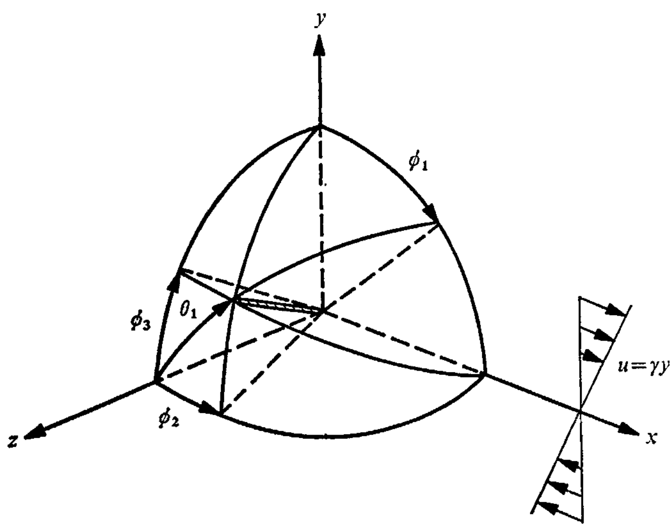

FIGURE 1. The co-ordinate system.

is identical, so far as rotation is concerned, to that of a spheroid with an effective axis ratio r_e which depends on the precise particle shape. This means that the results for spheroids which we shall present in subsequent sections can be adapted immediately to apply to any such body of revolution by simply substituting the effective axis ratio r_e for the spheroid axis ratio r.

The motion equations (2) can be integrated, with the result

$$\tan\phi_1 = r\tan\left(\frac{\gamma t}{r+(1/r)}\right), \tag{3a}$$

$$\tan\theta_1 = \frac{Cr}{(r^2\cos^2\phi_1+\sin^2\phi_1)^{\frac{1}{2}}}. \tag{3b}$$

According to these equations, the axis of revolution of a particle rotates about the z (vorticity) axis with a period

$$T = \frac{2\pi}{\gamma}\left(r+\frac{1}{r}\right) \tag{4}$$

and its ends describe a symmetric ellipse on the spherical surface

$$\left(\frac{x^2}{a^2}+\frac{y^2}{a^2}+\frac{z^2}{a^2}\right) = 1.$$

The constant of integration C is known as the orbit constant. As indicated in figure 2, it essentially defines the eccentricity of this orbit ellipse. We have already noted that the analysis leading to the orbit equations cannot be used to determine the orbit constant C for a single particle, there being no inherent preference for one orbit over any other within the framework of Jeffery's theory. The various proposals for dealing with this indeterminancy of C have been

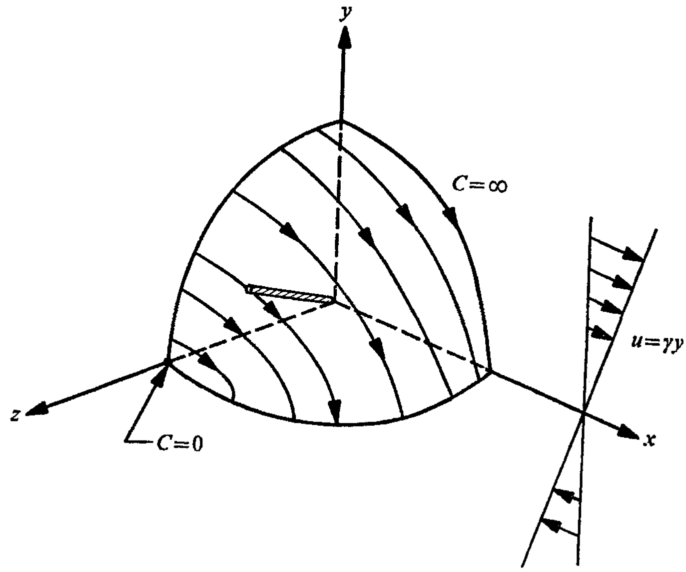

FIGURE 2. Typical orbits for slightly prolate spheroids ($r > 1$) in a shear flow.

described in detail in the previous section. Here, we consider the suggestion (Burgers 1938) that Brownian motion which is weak, though nevertheless dominant over inertial and particle-interaction effects, may produce a stationary distribution of orbits after a sufficiently long time.

We begin with the differential probability distribution function $N(\theta_1, \phi_1, t)$, defined such that the probability of finding any particular spheroid with its axis of revolution in the interval $[\theta_1, \theta_1 + d\theta_1] \times [\phi_1, \phi_1 + d\phi_1]$ on the unit sphere is

$$P(\theta_1, \phi_1, t) = N(\theta_1, \phi_1, t) \sin\theta_1 \, d\phi_1 d\theta_1. \tag{5}$$

Burgers (1938) has derived the general equation governing the time variations of the distribution function N, in the presence of Brownian motion, which is

$$\partial N/\partial t = -\operatorname{div}(\mathbf{w}N) + \operatorname{div}(D \operatorname{grad} N), \tag{6}$$

where D is the Brownian orientation 'diffusion' coefficient

$$D \equiv kT/R_w \tag{7}$$

and $$\mathbf{w}(\theta_1, \phi_1) \equiv (0, \dot{\theta}_1, \dot{\phi}_1 \sin\theta_1)$$

is the relative velocity (on the unit sphere) of the axis of revolution for a particle with instantaneous orientation, (θ_1, ϕ_1) ignoring all Brownian effects. In the expression for D, k is the Boltzmann constant ($1{\cdot}374 \times 10^{-16}$ erg/deg K), T is the absolute temperature in degrees Kelvin and R_w the resistance coefficient for rotation of the particle about a transverse axis through its centre, which for a spheroid is given by

$$R_w \equiv 4\mu V \left[\frac{r^2+1}{r^2 K_3 + K_1}\right], \tag{8}$$

where

$$K_3 \equiv \int_0^\infty \frac{r\,d\lambda}{(r^2+\lambda)^{\frac{3}{2}}(1+\lambda)}; \quad K_1 \equiv \int_0^\infty \frac{r\,d\lambda}{(r^2+\lambda)^{\frac{1}{2}}(1+\lambda)^2} \quad \text{and} \quad V \equiv \tfrac{4}{3}\pi a b^2.$$

The first term on the right-hand side of (6) represents the effect on N of motion around the Jeffery orbits. The second term represents the effect of random Brownian rotations, which introduce occasional small departures from the orientation distribution a particle would have if it were to rigorously follow a single Jeffery orbit. Because the sense of these Brownian motion orientation changes is random, they give rise to a diffusion-like process down gradients in the orientation probability space.

Solutions for equation (6) have previously been derived by a number of authors. Burgers himself considered the case of strong Brownian motion effects and computed the first few terms of an asymptotic series for N valid in the limit $D \gg 1$. Peterlin (1938) obtained a solution for N in terms of a slowly convergent series of spherical harmonics. Later, this series solution was numerically evaluated by Scheraga (1955) for values of D ranging from γ/D of zero to $\gamma/D = 60$, the latter representing the limit of storage capacity for the computer. In this paper we consider the solution to (6) in the limit of very weak Brownian diffusion.

3. Weak Brownian motion and its consequences

When the effect of Brownian diffusion is everywhere small, the steady-state distribution of orientations is governed by

$$\operatorname{div}(\mathbf{w}N) \equiv 0. \tag{9}$$

This equation is simply a statement that the particles are following the Jeffery orbits and yields no information about the relative populations of different orbits. Thus, without resort to an additional hypothesis regarding the initial state of the suspension (cf. Eisenschitz 1932), the complete neglect of all Brownian diffusion leaves an indeterminate problem for the probability distribution of orientations. The problem then is to deduce some condition in addition to the advection approximation (9) which will allow a determination of the distribution of orbit constants. Experience with the closely analogous problem of the steady distribution of vorticity for a closed streamline velocity field when the kinematic viscosity is very small (Batchelor 1956) indicates that the action of a small rate of Brownian diffusion over a sufficiently long time may eventually yield a determinate equilibrium distribution, N. Mathematically, this corresponds to the fact that the double limiting process involved in deriving equation (9) from equation (6) is not commutative and must be taken strictly in the order $t \to \infty$ and then $D \to 0$. Hence, we re-examine the original equation (6) and consider the consequences of the action of a small amount of Brownian diffusion in the limit as $t \to \infty$.

We begin by integrating (6) over a singly connected domain A of the unit sphere on which we have defined the function N,

$$\iint_A \frac{\partial N}{\partial t} dA = -\iint_A \operatorname{div}(\mathbf{w}N)\, dA + D\iint_A \operatorname{div}(\operatorname{grad} N)\, dA.$$

Applying the divergence theorem, we obtain

$$\iint_A \frac{\partial N}{\partial t} dA = -\oint (\mathbf{w}N).\mathbf{n}\, dl + D\oint (\operatorname{grad} N).\mathbf{n}\, dl,$$

where $\mathbf{n}$ is the outward normal of the bounding curve. Now choosing the bounding curve of the domain A to be a single Jeffery orbit, we find

$$\frac{\partial}{\partial t}\iint_A N\, dA = D\oint_C (\operatorname{grad} N).\mathbf{n}\, dl,$$

since $\mathbf{w}.\mathbf{n} = 0$. Hence, in a steady-state distribution, the net flux of particles across an orbit is zero,

$$D\oint_C \frac{\partial N}{\partial n} dl \equiv 0. \tag{10}$$

It is perhaps worth emphasizing that the condition (10) is independent of the value of D. In particular, it is applicable for arbitrarily small, though non-zero, values of the Brownian diffusion coefficient, provided only that as D becomes small other effects such as fluid inertia, and particle-particle interactions do not become of equal importance (in which case the original equation (6) would have to be modified). In the following section, we will employ (10) together with the advection equation (9) to determine the equilibrium distribution of orbit constants for situations in which the rate of Brownian diffusion across orbits is *everywhere* small compared to the advection around Jeffery orbits. Subsequent to obtaining this solution we will return to consider in detail the conditions necessary for equations (9) and (10) to be valid.

4. The equilibrium distribution of particle orientations in the limit of small Brownian diffusion

For convenience, we transform from the (θ_1, ϕ_1) co-ordinates on the unit sphere to the more natural orbit co-ordinates (C, τ). This transformation is essentially defined by the Jeffery orbit equations

$$\theta_1 = \tan^{-1}[C(\cos^2\tau + r^2\sin^2\tau)^{\frac{1}{2}}], \tag{11a}$$

$$\phi_1 = \tan^{-1}(r\tan\tau), \tag{11b}$$

where $\tau \equiv r\gamma t/(r^2+1)$. The co-ordinate lines C = const. and τ = const., though still on the unit sphere, are not orthogonal. We show typical co-ordinate lines in figure 3, together with the metrics of the system which we denote as h, k, and the angle α specifying the skewness of the co-ordinate lines. Hence,

$$ds^2 = d\theta_1^2 + \sin^2\theta_1 d\phi_1^2 = \theta_{1C}^2 dC^2 + 2\theta_{1C}\theta_{1\tau} dC\,d\tau + [\theta_{1\tau}^2 + \sin^2\theta_1 \phi_{1\tau}^2]\,d\tau^2, \quad (12)$$

where
$$\theta_{1C} \equiv \left(\frac{\partial\theta_1}{\partial C}\right)_\tau; \quad \theta_{1\tau} \equiv \left(\frac{\partial\theta_1}{\partial\tau}\right)_C; \quad \phi_{1\tau} \equiv \left(\frac{\partial\phi_1}{\partial\tau}\right)_C$$

and
$$\sin\theta_1 = \frac{C(\cos^2\tau + r^2\sin^2\tau)^{\frac{1}{2}}}{[1 + C^2(\cos^2\tau + r^2\sin^2\tau)]^{\frac{1}{2}}}.$$

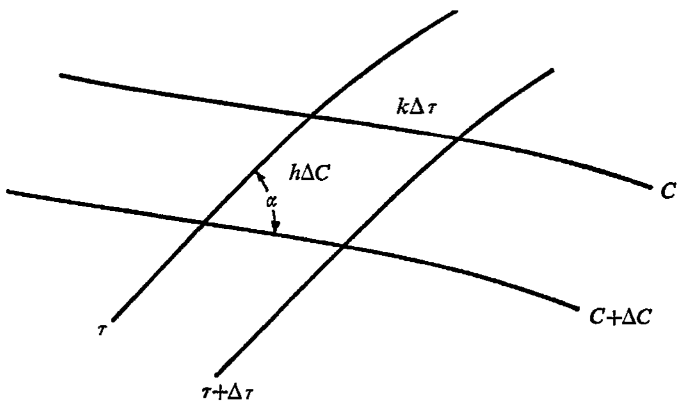

FIGURE 3. Typical co-ordinate lines for the orbit co-ordinates (C, τ).

Thus
$$k \equiv [\theta_{1\tau}^2 + \sin^2\theta_1\phi_{1\tau}^2]^{\frac{1}{2}}, \quad (13a)$$
$$h \equiv \theta_{1C} \quad (13b)$$
and
$$\sin\alpha \equiv \phi_{1\tau}\sin\theta_1[\theta_{1\tau}^2 + \sin^2\theta_1\phi_{1\tau}^2]^{-\frac{1}{2}}. \quad (13c)$$

Now, a first approximation to the probability distribution function N is given by the solution of equation (9) subject to the condition (10). (We will later see that this first approximation is uniformly valid provided that certain inequalities are satisfied.) In terms of the (C, τ) co-ordinates the equation (9) becomes

$$\frac{\partial}{\partial\tau}[N(C,\tau)\,h(C,\tau)\,k(C,\tau)\sin\alpha] = 0 \quad (14)$$

and hence,
$$N(C,\tau) = \frac{f(C)}{h(C_1\tau)\,k(C,\tau)\sin\alpha} = f(C)\,g(C,\tau). \quad (15)$$

Here, for convenience, we have denoted the group

$$[h(C,\tau)\,k(C,\tau)\sin\alpha]^{-1} \equiv g(C,\tau).$$

Thus, to a first approximation, the differential probability distribution function $N(C,\tau)$ is the product of a distribution around each of the various orbits, represented by $g(C,\tau)$, and a distribution across the orbits represented by the, as yet, unknown function $f(C)$. Employing (15) together with indicated co-ordinate transformation, the condition (10) for a stationary distribution of orientations can be expressed as

$$\int_0^{2\pi}\left[\frac{k}{h\sin\alpha}\frac{\partial}{\partial C} - \cot\alpha\frac{\partial}{\partial\tau}\right]f(C)\,g(C,\tau)\,d\tau \equiv 0, \quad (16a)$$

or equivalently

$$\left[\int_0^{2\pi} \frac{d\tau}{h^2 \sin^2 \alpha}\right] \frac{df}{dC} + \left[\int_0^{2\pi} \left\{\frac{k}{h \sin \alpha} \frac{\partial g}{\partial C} - \cot \alpha \frac{\partial g}{\partial \tau}\right\} d\tau\right] f = 0. \tag{16b}$$

Evaluating the integrals in (16*b*), we obtain the variable coefficient, ordinary differential equation for f,

$$O \equiv [H(r)\, C^4 + K(r)\, C^2 + M(r)] \frac{df}{dC} + \frac{1}{C}[2H(r)\, C^4 + (6 - K(r))\, C^2 - M(r)] f, \tag{16c}$$

where

$$H(r) \equiv r^2 + 1;\ K(r) \equiv \tfrac{1}{4}r^2 + \tfrac{7}{2} + 1/4r^2 \quad \text{and} \quad M(r) \equiv (r^2+1)/r^2.$$

This equation yields the general solution

$$f(C, r) = \text{const.}\, C[(HC^4 + KC^2 + M)\, F(C, r)]^{-\frac{3}{4}}, \tag{17}$$

in which

$$F(C, r) \equiv \left[\frac{2HC^2 + K - (K^2 - 4HM)^{\frac{1}{2}}}{2HC^2 + K + (K^2 - 4HM)^{\frac{1}{2}}}\right]^{(4-K)/(K^2-4HM)^{\frac{1}{2}}} \qquad (K^2 > 4HM);$$

$$F(C, r) \equiv \exp\left[\frac{2(K-4)}{2HC^2 + K}\right] \quad (K^2 = 4HM);$$

$$F(C, r) \equiv \exp\left[\frac{2(4-K)}{(4HM - K^2)^{\frac{1}{2}}} \tan^{-1} \frac{2HC^2 + K}{(4HM - K^2)^{\frac{1}{2}}}\right] \quad (K^2 < 4HM).$$

The constant of integration is found by normalizing the probability density function

$$1 = \int_{\text{unit sphere}} \frac{N(C, \tau)}{g(C, \tau)}\, dC\, d\tau,$$

or

$$\int_0^{\infty} f(C)\, dC = 1/4\pi. \tag{18}$$

The orientation distribution function $N(C, \tau)$ follows directly from (17), (18) and (15). Although we have not been able to analytically integrate the function f to obtain the integration constant for arbitrary values of the particle aspect ratio r, the three limiting cases $r = 1$, $r \to \infty$ and $r \to 0$ all allow analytical normalization. The resulting expressions are

$$f(C) = \frac{1}{4\pi} \frac{C}{(C^2+1)^{\frac{3}{2}}} \quad (r = 1) \tag{19a}$$

$$f(C, r) \sim \frac{1}{\pi} \frac{C}{(4C^2+1)^{\frac{3}{2}}} \quad (r \to \infty) \tag{19b}$$

and

$$f(C, r) \sim \frac{1}{\pi} \frac{Cr^2}{(4r^2C^2+1)^{\frac{3}{2}}} \quad (r \to 0), \tag{19c}$$

respectively. In the general case, we have performed the normalization numerically. The resulting solution has been plotted in figure 4 for various values of r in the range ($0{\cdot}01 \leqslant r \leqslant 1000$). Although we have not separately plotted the asymptotic results (19), we note that for $r < 20$, and $r < 0{\cdot}05$ the numerical and asymptotic results are nearly identical. For comparison, we have also plotted the function f evaluated using the Eisenschitz hypothesis. Looking first at our numerically normalized solution, we note that as r is increased from 1, there are proportionally more particles in orbits with low values of C whereas when r is decreased from 1, the opposite effect is observed. Indeed, according to (19*b*), the distribution function f has a maximum value $1/3^{\frac{3}{2}}\pi$ at a value of C equal to $1/2^{\frac{1}{2}}$ when $r \gg 1$. On the other hand, the maximum for $r \ll 1$ occurs for C equal to $1/2^{\frac{1}{2}}r$ and only has a magnitude of $r/3^{\frac{3}{2}}\pi$. Both of these asymptotic characteristics

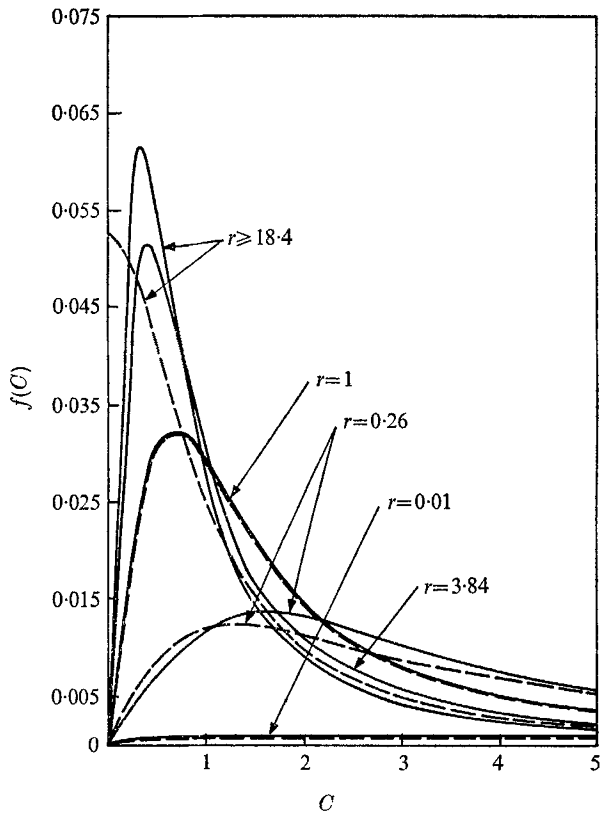

FIGURE 4. The orbit distribution function for various particle axis ratios: ——, our theory; - – – –, Eisenschitz hypothesis.

are evident in the numerically normalized solutions of figure 4. Comparing these normalized solutions with the equilibrium distribution which is calculated from the Eisenschitz hypothesis, it is striking that the two are very similar except near $C = 0$ where the latter are generally somewhat larger.

The distribution function $f(C)$ for $r = 1$ is of particular interest since it allows a limited test of the internal consistency of our theory. A spherical particle simply rotates about the vorticity vector with a rate of rotation $\frac{1}{2}\gamma$. Hence any arbitrary axis passing through the centre of the particle traces out a circle about the vorticity axis. Furthermore, in this degenerate case, *any* axis can be chosen as the axis of revolution. Thus the probability distribution function for the orientation of this axis will be uniform in ϕ_1, and vary with θ_1 as $\sin\theta_1$. It may be simply shown that the distribution function $f(C)$ (equation (19a)) produces this expected result.

Before turning to considerations of the range of validity of the equations (9) and (10) and hence of our solution (17), it is instructive to consider the qualitative implication of our solution with regard to the actual *orientation* of the particles. In order to do so, however, it is convenient to consider first the nature of the *possible* orbits. Since the transformation

$$r \to 1/r; \quad \theta_1 \to \theta_1; \quad \phi_1 \to \phi_1 \pm \tfrac{1}{2}\pi$$

leaves the orbit equations invariant, we shall primarily consider prolate spheroids. For simplicity we shall further focus our discussion on the case $r \gg 1$, corresponding to slender rods. We have illustrated a set of typical orbits for a moderate value of $r(>1)$ in figure 2. We note that the limiting orbits $C = 0$ and ∞ are independent of the particle aspect ratio r, corresponding, respectively, to the axis of revolution being exactly aligned with the vorticity vector of the basic shear flow ($C = 0$) and the particle rotating completely in the plane of the shear flow ($C = \infty$). The shape of all the remaining orbits ($0 < C < \infty$) depends on r. In the limit as $r \to \infty$, the orbit equations (3a, b) can be approximated by

$$\tan\phi_1 \sim \gamma t; \quad \tan\theta_1 \cos\phi_1 \sim C,$$

provided only that $|\phi_1 - \frac{1}{2}\pi| > O(1/r)$. Employing the angle transformation $\tan\theta_1 \cos\phi_1 = \tan\phi_3$, the latter becomes

$$\tan\phi_3 \sim C \quad \text{or} \quad \phi_3 \sim \tan^{-1} C = \text{const.}$$

The angle ϕ_3 is defined in figure 1. Hence, in the limit as $r \to \infty$, *all* of the possible orbits follow a path $\phi_3 \sim$ const., thus eventually approaching the plane of zero flow (ZX) and hence passing into the region $|\phi_1 - \frac{1}{2}\pi| \leqslant O(1/r)$. Within this region, the rate of change of ϕ_1 becomes very small compared to its value elsewhere in the orbit, i.e.

$$\dot{\phi}_1 \sim O(\gamma/r^2)$$

and the orbit itself deviates from $\phi_3 \sim$ const. to a path which becomes normal to the ZX plane as $\phi_1 \to \frac{1}{2}\pi$ and crosses it at an angle θ_1^* which, from (3b), is

$$\theta_1^* = \tan^{-1}(Cr).$$

Provided $C \geqslant O(1/r)$ this angle $\theta_1^* \sim \frac{1}{2}\pi$, and hence, a particle spends a great deal of its period essentially aligned with the flow. Now according to our asymptotic evaluation of the distribution $f(C, r)$, equation (19b), when $r \gg 1$ this description is valid for the majority of particles since most end up in orbits for which C is $O(1)$. The exceptional orbits where this description is not accurate are those for which $C \leqslant O(1/r)$ and is hence very small. In these cases, the particle crosses the ZX plane at an angle θ_1^* which is not near $\frac{1}{2}\pi$ and the angle $\phi_3 \leqslant O(1/r)$ everywhere on the orbit.

A physical explanation for the calculated distribution of orbits is as follows. When the suspension is initially subjected to the shear flow, the majority of particles are presumably in orbits with $C > O(1/r)$ and hence, according to our previous discussion, they spend most of their time nearly lined up with the flow. Thus, initially the orientation distribution function has a sharp maximum for $\phi_1 \sim \theta_1 \sim \frac{1}{2}\pi$ and is nearly zero everywhere else. The rotational Brownian diffusion tends to spread this initial concentration distribution more evenly over the unit sphere, and since all orbits cause the particles to spend most of their time very near the ZX plane this initial diffusion would, in the absence of competing effects, tend primarily to produce a more uniform distribution over θ_1^* in the range 0 to $\frac{1}{2}\pi$. However, orbits yielding intermediate values of θ_1^* correspond to $C \leqslant O(1/r)$ and we have already seen that $\phi_3 \leqslant O(1/r)$ everywhere for these orbits. Hence the tendency to establish a uniform distribution over θ_1^* is counteracted by Brownian diffusion resulting from increased gradients of N in the ϕ_3 direction. It is clear, upon examining the asymptotic distribution, equation (19*b*), that the effect of diffusion in the ϕ_3 direction largely dominates the tendency toward a uniform distribution over θ_1^*. The resulting equilibrium distribution of orientations is still very much concentrated near $\theta_1^* = \frac{1}{2}\pi$. This apparent dominance of the effect of diffusion across ϕ_3 is certainly not surprising since very small fluctuations of orientation over ϕ_3 (outward from the ZX plane) will tend to result in large changes in the angle θ_1^* back toward the value $\frac{1}{2}\pi$. The transfer of particles *into* the orientation region $\phi_3 \leqslant O(1/r)$, which would have the opposite effect on θ_1^*, will be much less effective since the residence time over the portions of the orbit away from the ZX plane is a very small fraction of the total orbit period. Due to symmetry considerations, a mirror image distribution is established for $\phi_1 \sim -\frac{1}{2}\pi$.

Considering the transformation from rods to disks cited earlier, we would expect the distribution for disks to show an initially strong concentration of orientations near $\phi_1 = 0$ (and π) $\theta_1 = \frac{1}{2}\pi$ with the tendency toward a redistribution over $\theta_1|_{\phi_1=0}$ largely dominated by the tendency of diffusion to diminish the resulting large gradients in N with respect to ϕ_2.

5. The conditions for validity for our theory

Before calculating various bulk properties for a dilute suspension of spheroids in the presence of the weak Brownian motion effects discussed in this paper, it is of some interest to examine more carefully the conditions necessary for the equations (9) and (10) to be valid. First of all, we consider the condition for Brownian diffusion to be dominated everywhere in orientation space by the advection of particles around Jeffery orbits. That is, we wish to determine the conditions such that

$$\left\| \frac{\partial}{\partial x_j} w_i N \right\| \gg D . \left\| \frac{\partial^2 N}{\partial x_i \partial x_j} \right\|$$

everywhere on the unit sphere, so that (9) is uniformly valid, and hence the resulting solution (15) can be considered as the first term of a *regular* perturbation expansion for N. We define a suitable local Péclet number for Brownian diffusion of N as

$$P \equiv UL/D,$$

where U, L are characteristic length, $|\nabla \log N|^{-1}$, and velocity $|\mathbf{w}|$, scales in the orientation space and D is the Brownian diffusion coefficient. We require that the *local* Péclet number be everywhere large compared to one, i.e.

$$P_{\min}^{-1} \ll 1,$$

where $P_{\min}$ denotes the smallest value of the local Péclet number on the unit sphere.† The minimum *local* values of velocity and length scale for an orbit occur at $\phi_1 = \frac{1}{2}\pi$ for rods and $\phi_1 = 0$ for disks, and can be estimated from equations (2), (15) and (19) for these limiting cases as

$$U_{\min} \sim \gamma/r^2 \quad \text{and} \quad L_{\min} \sim 1/r \quad (r \to \infty).$$

$$U_{\min} \sim \gamma r^2 \quad \text{and} \quad L_{\min} \sim r \quad (r \to 0).$$

Hence, to ensure the validity of (9), we require

$$Dr^3/\gamma \ll 1 \quad (r \to \infty) \quad \text{and} \quad D/\gamma r^3 \ll 1 \quad (r \to 0) \tag{20a}$$

respectively.

Expanding D (equation (7)) asymptotically for large and small r, these become

$$\frac{kTr(r^2K_3+K_1)}{4\mu\gamma V} \ll 1 \quad (r \to \infty); \quad \frac{kT(r^2K_3+K_1)}{4\mu\gamma Vr^3} \ll 1 \quad (r \to 0). \tag{20b}$$

For a given value of r, these conditions can be satisfied by making the product of the particle volume, the suspending fluid viscosity (μ), and the rate of shear (γ) sufficiently large, or by making the temperature sufficiently small.

In addition to these conditions, we must also require that in spite of Brownian diffusion being small, it still remains dominant over the effects of inertia and particle-particle interactions so far as the change in orbit constant is concerned. Unfortunately, neither the inertia nor the interaction problem has yet been totally resolved for spheroidal particles so that it is not entirely obvious what the appropriate parameters for their neglect in our work should be. The physical requirement is that the average rate of change in C for a particle be small compared to that caused by Brownian rotations. We believe that requiring the overall particle Reynolds number (based on the maximum linear dimension (i.e. a or b) of the particle and the bulk shear rate γ) to be sufficiently small is equivalent to this physical requirement in the inertia case, so that one condition is simply

$$\frac{[\max(a,b)]^2\gamma}{\nu} \ll P_{\min}^{-1}. \tag{21}$$

Intuitively, one expects the effect of particle interaction (on the average change in C) to be small compared to the Brownian motion effect for some sufficiently small volume concentration Φ of suspended particles. To obtain a quantitative estimate of the importance of particle interactions, we *conjecture* that the effect of interactions on the rate of change in C is sensibly measured by the ratio of the effective volume occupied by a particle ($\frac{4}{3}\pi a^3$ for rods, $\frac{4}{3}\pi b^3$ for disks) to the average volume available to each particle of the suspension (i.e. total volume/number of particles). This assumption is at least consistent with the intuitive motion that the magnitude of the interaction effect will be estimated by the average probability of finding any one particle in the disturbed velocity field of another. On

† Although it is not possible to use the heuristic arguments employed here to rigorously justify the exponent (-1) in this inequality, we note that the resulting conditions (equations (20a, b)) are precisely the same as those obtained from very recent work based on calculating higher-order corrections to the orientation distribution function.

this basis, an order of magnitude of the condition for neglect of particle interactions is

$$O\left(\frac{\text{effective volume}}{\text{average volume}}\right) = \Phi r^2 \ll P_{\min}^{-1} \quad (r \to \infty), \tag{22a}$$

$$= \Phi / r \ll P_{\min}^{-1} \quad (r \to 0). \tag{22b}$$

Clearly this requirement implies that the volume concentration Φ of suspended particles must be very small indeed, especially in respective limits as $r \to \infty$ or $r \to 0$.

6. The probability distributions of $(\phi_2)_{\max}$ and $(\phi_3)_{\max}$

Although we have already discussed qualitatively the equilibrium distribution of particle orientations in the limits $r \to \infty$ and $r \to 0$, we shall, nevertheless, derive here some quantitative results for the distribution over the maximum values (attained during an orbit) of the angles ϕ_2 and ϕ_3. The primary reason for interest in these maximum angle distributions is that they are, in principle, conveniently measured quantities which have been purported to provide a reasonably sensitive determination of the probability distribution of orbits.

We have defined the angles ϕ_2 and ϕ_3 in figure 1. When projected on the (ZX) plane the motion of a particle appears as a rocking of the axis of revolution back and forth between maximum angles $\pm(\phi_2)_{\max}$. Now it can be shown from Jeffery's equations that

$$\tan \phi_2 = Cr \sin(2\pi t/T)$$

in which T is the orbital period defined in (4). Thus, the amplitude of the rocking motion, projected on the (ZX) plane, is directly related to the orbit constant C of the particle, and, in fact, the maximum value of ϕ_2 is given by

$$(\phi_2)_{\max} = \tan^{-1}(Cr). \tag{23}$$

Hence the measurement of $(\phi_2)_{\max}$ for particles of known axis ratio is equivalent to a determination of the orbit constant C. We define $P(\phi_{2m})$ as the probability of a particle having $(\phi_2)_{\max}$ between zero and ϕ_{2m}. Clearly, in view of (23), an equivalent definition of $P(\phi_{2m})$ is the probability of a particle having an orbit constant C between 0 and $(1/r)\tan\phi_{2m}$. In the latter form, $P(\phi_{2m})$ can be calculated simply from the distribution function $f(C)$, equations (17)–(18), by the relation

$$P(\phi_{2m}) = 4\pi \int_0^{(1/r)\tan\phi_{2m}} f(C')\,dC'. \tag{24}$$

We have evaluated this expression for various values of r in the range 10^{-2}–10^3 using the numerically normalized distribution function $f(C)$, and the results are shown in figure 5. For comparison, we have also calculated the probability function, $P(\phi_{2m})$, first using the asymptotic evaluations of $f(C)$ (equations (19b, c)), and second, the Eisenschitz distribution of orbit constants. The latter are shown plotted in figure 5. The former give

$$P(\phi_{2m}) \sim \left[1 - \frac{r}{(4\tan^2\phi_{2m} + r^2)^{\frac{1}{2}}}\right] \quad (r \to \infty), \tag{25a}$$

$$P(\phi_{2m}) \sim \left[1 - \frac{1}{(4\tan^2\phi_{2m} + 1)^{\frac{1}{2}}}\right] \quad (r \to 0) \tag{25b}$$

and these expressions agree well with the 'numerical' values plotted in figure 5 for $r < 20$ and $r > 0{\cdot}05$, respectively. Also shown in figure 5 are some measured distributions of $P(\phi_{2m})$ due to Anczurowski & Mason (1967). Clearly, there is a considerable similarity between our predicted distribution and the Eisenschitz distribution. On the other hand, a considerable difference is evident between both of these, and the experimentally measured distributions of Anczurowski & Mason (1967). We will return to consider the possible implications of this difference.

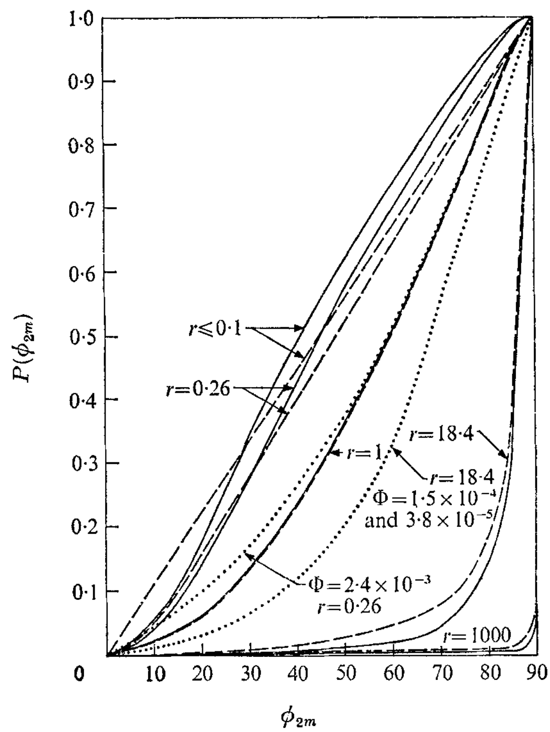

FIGURE 5. The probability function $P(\phi_{2m})$ for various particle axis ratios: ——, our theory; – – – –, Eisenschitz hypothesis; ..., experiment (Anzcurowski & Mason 1967), Φ is volume concentration of particles.

In order to determine the probability function for $(\phi_3)_{\max}$, we note the following relationship between the distribution functions $P(\phi_{2m})$ and $P(\phi_{3m})$,

$$P(\phi_{2m})|_{r=R} = P(\phi_{3m})|_{r=1/R}. \tag{26}$$

This is, of course, simply a reflexion of the symmetries in the equilibrium distribution of orientations which we discussed in the previous section. Clearly, the distribution functions $P(\phi_{3m})$ predicted by our theory can be read directly from figure 5. We note that an experimental determination of $P(\phi_{3m})$ is rather difficult since, in order to measure $(\phi_3)_{\max}$, it is necessary to view the particle along lines *parallel* to the flow. Hence, so far as we are aware, no measurements of $P(\phi_{3m})$ have yet been reported.

In principle, neither $P(\phi_{2m})$ nor $P(\phi_{3m})$ is an entirely satisfactory means of determining the distribution of orbit constants, since each is insensitive to a portion of this distribution in the limiting cases of small and large r. For examples the distribution $P(\phi_{2m})$ fails to distinguish between orbits with $C > 1/r$ when $r \gg 1$. The explanation for this insensitivity is that *all* orbits with $C > 1/r$ pass very near the point $\theta_1 = \phi_1 = \frac{1}{2}\pi$, this corresponding to $(\phi_2)_{\max} = \frac{1}{2}\pi$. A similar insensitivity of $P(\phi_{3m})$ to C arises when $r \ll 1$. Hence, if we could ensure that the majority of particles in a suspension had orbit constants of $O(1)$ and $O(r)$, respectively, then a reasonable plan would be to measure $P(\phi_{3m})$ for $r > 1$ and $P(\phi_{2m})$ for $r < 1$. Unfortunately, however, if $C = O(1/r)$ as $r \to \infty$ or $O(1)$ as $r \to 0$ (as is clearly possible), then the optimum distribution function for determining C would be just the opposite of those for the 'general' case. Thus $P(\phi_{2m})$ would be most satisfactory for $r > 1$ whereas $P(\phi_{3m})$ would work best for $r < 1$. Clearly then, when r is extreme, there can be serious difficulties inherent in attempting to deduce the orbit constant distribution from experimental measurements of either $P(\phi_{2m})$ or $P(\phi_{3m})$. We do not believe, however, that the poor comparison (figure 5) between our predicted distributions and the measured ones of Anczurowski & Mason (1967) can be attributed, to any significant extent, to these possible difficulties since, for the most part, the particle aspect ratio in the experiments was neither very large nor very small. Rather, we believe that the discrepancy is due to *real* differences in the orbit distribution functions for the two cases. Now the measured distributions were indicated by Anczurowski & Mason (1967) as being determined primarily by particle-particle interactions. On this basis, we can conclude that even in the rather dilute suspensions employed by these authors ($\Phi \sim 10^{-4}$) it is not possible to model the effect of particle interactions as small random changes in orientation. Otherwise the theoretical distribution functions determined here should have been the same as the experimental ones, in spite of the obvious differences in the underlying mechanism. Intuitively one might have supposed that distant interactions would be dominant over the much less frequent but strong close-particle interactions, and further, that these distant interactions could be modelled by small random changes of orientation. Since our present results seemingly exclude such a model, it must be concluded that either distant interactions are not adequately represented by weak random rotations or else that the effect of near interactions simply cannot be neglected in any circumstances where particle-particle interactions are important (or perhaps both).

7. The effective viscosity

The suspensions which we have considered are anisotropic, and so in general the bulk stress-bulk rate of strain relation will not be Newtonian in form (Batchelor 1970). However, the non-Newtonian nature of the stress in a simple shear flow (normal stress differences) will be small relative to the Newtonian type contributions in the limiting situation which we have considered. Hence in the interest of simplicity we have elected to consider only the ratio of the tangential stress to the rate of shear γ and we designate this as an effective viscosity, μ^*.

Jeffery (1932) derived an expression for the effective viscosity of a dilute suspension of spheroids which is accurate in the limit of weak Brownian rotational diffusion and is given by

$$\frac{\mu^*-\mu}{\mu\Phi} = \langle\sin^4\theta_1\sin^2 2\phi_1\rangle\left\{\frac{J_3}{I_3J_2}+\frac{1}{I_3}-\frac{2}{I_2}\right\}+\langle\cos^2\theta_1\rangle\frac{2}{I_3}+\langle\sin^2\theta_1\rangle\frac{2}{I_2}. \tag{27}$$

The underlying assumption inherent in this expression is that the contribution of Brownian motion is, at least to first-order accuracy, entirely manifested in its effect on the orientation distribution of the suspension particles. Here I_2, I_3, J_2, J_3, are non-dimensional forms of Jeffery's particle shape functions and are defined, for the general ellipsoid, by Batchelor (1970.) The notation $\langle\,\rangle$ indicates an average over all the particles of the supension. Jeffery also gave expressions for the angle averages over a given orbit, C, which are

$$\langle\sin^4\theta_1\sin^2 2\phi_1\rangle_C = \frac{2r^2}{(r^2-1)^2}\left\{\frac{C^2(r^2+1)+2}{[(C^2r^2+1)(C^2+1)]^{\frac{1}{2}}}-2\right\}, \tag{28a}$$

$$\langle\cos^2\theta_1\rangle_C = \frac{1}{[(C^2r^2+1)(C^2+1)]^{\frac{1}{2}}}. \tag{28b}$$

Hence in the case of a given (or known) distribution function over all possible orbits, $f(C)$, the averages over all the particles in the suspension are simply

$$\langle\sin^4\theta_1\sin^2 2\phi_1\rangle = \int_0^\infty \langle\sin^4\theta_1\sin^2 2\phi_1\rangle_C\, 4\pi f(C)\,dC \tag{29a}$$

and

$$\langle\cos^2\theta_1\rangle = \int_0^\infty \langle\cos^2\theta_1\rangle_C\, 4\pi f(C)\,dC. \tag{29b}$$

The factor 4π arises because of our choice of normalization for $f(C,r)$.

We have evaluated the expressions (29) and thence the effective viscosity (27), using the definitions (28) and our numerically normalized distribution function $f(C,r)$. The results are shown in table 1. We note the expected symmetric character of the angle averages with respect to the transformation $r \to 1/r$.

The expressions (29) can also be determined from the asymptotic forms of $f(C)$, equations (19*b*) and (19*c*). The angle averages become

$$\left.\begin{aligned}\langle\sin^4\theta_1\sin^2 2\phi_1\rangle &\sim 1{\cdot}248/r\\ \langle\cos^2\theta_1\rangle &\sim 1{\cdot}792/r\end{aligned}\right\}\quad r\to\infty, \tag{30a}$$

$$\left.\begin{aligned}\langle\sin^4\theta_1\sin^2 2\phi_1\rangle &\sim 1{\cdot}248r\\ \langle\cos^2\theta_1\rangle &\sim 1{\cdot}792r\end{aligned}\right\}\quad r\to 0. \tag{30b}$$

The symmetry noted previously (for $r\to 1/r$) in the numerically evaluated results is clearly evident here. Asymptotic evaluation of the quantities I_2, I_3, J_2, J_3 for spheroids symmetric about their 3 axis gives

$$\left\{\frac{J_3}{I_3J_2}+\frac{1}{I_3}-\frac{2}{I_2}\right\}\sim\frac{1}{4}\frac{r^2}{\ln 2r-1{\cdot}5};\quad \frac{2}{I_3}\sim 2;\quad \frac{2}{I_2}\sim 2\quad (r\to\infty)$$

and

$$\left\{\frac{J_3}{I_3J_2}+\frac{1}{I_3}-\frac{2}{I_2}\right\}\sim\frac{5}{3\pi r};\quad \frac{2}{I_3}\sim\frac{8}{3\pi r};\quad \frac{2}{I_2}\sim 1\quad (r\to 0).$$

r	$\langle \sin^4\theta_1 \sin^2 2\phi_1\rangle$	$\langle \cos^2\theta_1\rangle$	$\langle \sin^4\theta_1 \sin^2 2\phi_1\rangle$ equation (30*a*)	$\langle \sin^4\theta_1 \sin^2 2\phi_1\rangle$ equation (30*b*)	$\langle \cos^2\theta_1\rangle$ equation (30*a*)	$\langle \cos^2\theta_1\rangle$ equation (30*b*)	$\frac{\mu^*-\mu}{\mu\Phi}$	$\frac{\mu^*-\mu}{\mu\Phi}$ equation (31*a*)	$\frac{\mu^*-\mu}{\mu\Phi}$ equation (31*b*)
1000	0·00125	0·00174	0·00125	—	0·00179	—	50·062	49·990	—
100	0·01217	0·01727	0·01248	—	0·01792	—	10·008	10·213	—
20	0·05425	0·07921	0·06240	—	0·08960	—	4·403	4·851	—
18.4	0·05828	0·08509	0·06783	—	0·09739	—	4·277	4·726	—
10	0·09570	0·13820	0·12480	—	0·17920	—	3·464	4·086	—
3·846	0·17995	0·24614	0·32449	—	0·46594	—	2·784	4·222	—
2·440	0·22193	0·29329	0·51148	—	0·73443	—	2·618	10·956	—
1·05	0·26361	0·33729	—	—	—	—	2·500	—	—
0·41	0·22193	0·29329	—	0·51148	—	0·73443	2·603	—	2·448
0·26	0·17995	0·24614	—	0·32449	—	0·46594	2·712	—	2·717
0·10	0·09570	0·13820	—	0·12480	—	0·17920	2·952	—	3·004
0·05	0·05425	0·07920	—	0·06240	—	0·08960	3·071	—	3·093
0·01	0·01218	0·01726	—	0·01248	—	0·01792	3·121	—	3·165
0·001	0·00125	0·00174	—	0·00125	—	0·00179	3·140	—	3·181

TABLE 1

Hence, the corresponding asymptotic expressions for the effective viscosity are

$$\frac{\mu^*-\mu}{\mu\Phi} \sim 2+\frac{0\cdot 312r}{\ln 2r-1\cdot 5} \quad (r\to\infty) \tag{31a}$$

and

$$\frac{\mu^*-\mu}{\mu\Phi} \sim 3\cdot 183-1\cdot 792r \quad (r\to 0). \tag{31b}$$

Table 1 shows a comparison between the numerically evaluated angle averages and effective viscosity and the asymptotic evaluations using equations (30) and (31). For $r > 20$ and $< 0\cdot 05$ respectively, the two sets of values agree well. The effective viscosity increases rapidly as r increases from the spherical value ($r = 1$). On the other hand, the effective viscosity for a suspension of disks decreases as r is decreased, the expression $(\mu^*-\mu)/\mu\Phi$ asymptotically approaching the value 3·183 as $r\to\infty$.

Although a number of measurements of suspension viscosities for rods and disks have been reported they have all involved volume concentrations which are too large for our theory to be valid. This is primarily due to the large size of particles employed. With smaller particles, a relatively large number density could be allowed without violating any of our conditions (20), (21), or (22). It would be particularly interesting to determine whether measured viscosities of such a suspension would agree with those of table 1, since, especially in the limiting case as $r\to\infty$, the effective viscosity is very sensitive to small changes in the distribution of the orientations, and would thus offer a sensitive initial check on the validity of our theory.

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