Particle shapes leading to Newtonian dilute suspensions

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(Received 16 December 2019; accepted 1 September 2020; published 25 September 2020)

It is well known that suspensions of particles in a viscous fluid can affect the rheology significantly, producing a pronounced non-Newtonian response even in dilute suspension. However, it is unclear *a priori* which particle shapes lead to this behavior. We present two simple symmetry conditions on the shape which are sufficient for a dilute suspension to be Newtonian for all strain sizes and one sufficient for Newtonian behavior for small strains. We also construct a class of shapes out of thin, rigid rods not found by the symmetry argument which share this property for small strains.

DOI: 10.1103/PhysRevE.102.032615

I. INTRODUCTION

The theoretical study of rigid particle suspensions dates back to Einstein's seminal work [1,2]. He showed that introducing a dilute suspension of spheres to a viscous solvent increased the solvent's viscosity from η_s to $\eta_s(1 + 2.5n\nu)$, where n is the number density and ν is the volume of each sphere. Since then, a wide variety of systems have been successfully modeled, from polymeric fluids [3] to active bacterial suspensions [4].

The rheology of a fluid is understood through the constitutive equation relating the stress, σ, and the rate of strain tensor κ. For a Newtonian fluid, the stress is linearly related to the strain by a time independent 4th rank tensor. For isotropic, incompressible fluids this tensor is isotropic and determined solely by the fluid's viscosity. For small strains, the stress in a general fluid is characterized by the complex viscosity, $\eta^*(\omega)$. This is defined such that the stress response to the oscillatory shear, $\kappa_{xy} = \dot{\gamma}\,\mathrm{Re}(e^{i\omega t})$, is given by $\sigma_{xy} = \dot{\gamma}\,\mathrm{Re}[\eta^*(\omega)e^{i\omega t}]$, where $\dot{\gamma}$ is the shear rate and ω is the angular frequency. The real and imaginary parts of η^* represent the viscosity and elasticity of the solution, respectively, both of which may be dynamic. A fluid whose complex viscosity has both real and imaginary parts is viscoelastic. The complex viscosity of a Newtonian fluid is a real constant, independent of frequency. Our results do not depend on the form of κ. We used oscillatory shear as an illustrative example, as oscillatory rheology is a common experimental technique.

The presence of particles can change the behavior of a fluid, producing a non-Newtonian response. While the most extreme phenomena occur at large concentrations or strains, it has been shown that there exists a small viscoelastic effect in dilute suspensions of certain particle shapes. This is not the case for spheres. Notably, when their Brownian motion is taken into account, rigid rodlike particles show a finite, linear elastic response in dilute suspension [3]. Other particle shapes, such as spheroids [5] and propellerlike particles [6], have also been shown to behave similarly. Clearly, the presence of elasticity in these suspensions depends on the particles' shape.

The magnitude of this effect is proportional to the concentration of particles, and is therefore very small in dilute suspension. However, it is possible to measure this effect through the intrinsic complex viscosity [3,7], defined as

$$[\eta^*(\omega)] = \lim_{n\to 0} \frac{\eta^*(\omega) - \eta_s}{n\eta_s}. \tag{1}$$

If the suspension is Newtonian, then the intrinsic viscosity will be independent of frequency.

While the methods for finding the stress in such systems are well known [8,9], it is unclear *a priori* which particle shapes will lead to a non-Newtonian response, without simulations or cumbersome calculations.

In this paper, we aim to understand and characterize the properties that a particle shape must have for a dilute suspension to be Newtonian. We determine two simple symmetry conditions on the particle shape which, when both are satisfied, are sufficient for the suspension to be Newtonian. These are derived by considering the symmetries of the particle, without referencing its specific shape. As long as the particles' positions remain uniformly distributed and interparticle interactions are negligible, these hold for all strain sizes. In the case of small strains, one of these two conditions is relaxed, and there is only one sufficient condition for purely viscous behavior. Examples of particle shapes with Newtonian dilute suspensions are shown in Fig. 1(a); the underlined shapes have a Newtonian response for all strain sizes. The conditions presented here make this identification straightforward where explicit calculations would be extremely difficult, e.g., for the Archimedian solids.

The ability to predict the presence of a non-Newtonian response for particles with arbitrary shapes has become highly relevant given the emergence of sophisticated techniques [10,11] to design the shape of nanoparticles. We specifically reference DNA nanostars [12–14], constructed from linked double stranded DNA sequences, which have recently been

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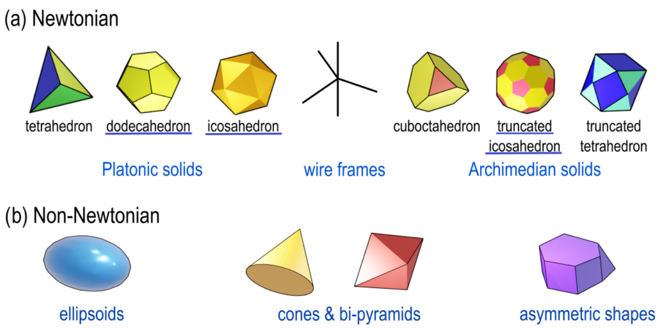

FIG. 1. (a) Examples of particle shapes which have Newtonian dilute suspensions for small strains. Due to their symmetry, the Platonic and Archimedean solids produce no elasticity. Some wire-frame shapes share this property if the ratios of the rod lengths are chosen correctly. The underlined shapes lead to Newtonian suspensions for all strain sizes. (b) Examples of particle shapes which, generally, lead to non-Newtonian dilute suspensions.

synthesized and studied for their potential biomedical and nanoengineering applications.

We provide a general method for describing the rheology of dilute suspensions of these particles, based on the Onsager Principle [15,16], a powerful tool for describing the behavior of a wide range of systems [17,18]. We use this approach to demonstrate the predictions of the symmetry argument for small strains for specific shapes, and construct a class of shapes not found by the symmetry argument whose dilute suspensions also have a purely viscous linear response.

Our results are only valid in dilute suspension where the effect of the particles is very small, and therefore less important for practical applications. Nevertheless, they indicate something fundamental about the particles' influence on the viscoelastic properties of the fluid. For particular particle shapes, all non-Newtonian behavior is due to interparticle interactions.

II. OVERVIEW FOR GENERAL SHAPES

We begin with a brief overview of the method for determining the viscoelasticity of a dilute suspension of particles of general shape; a full description may be found in Makino and Doi [6].

There are two contributions to the stress in these systems: one arising purely from the hydrodynamics of the suspended particles and the other from their Brownian motion. We assume that the particle sizes, velocity, and viscosity of the fluid are such that the Reynolds number may be taken to be small. To describe the hydrodynamics in this regime, we only need to consider one particle whose linear velocity, $\mathbf{v}$, and angular velocity, $\mathbf{\Omega}$, are linearly related to those of the fluid and the rate of strain tensor of the flow via mobility matrices which depend on the geometry and orientation of the particle [8,9].

The Brownian motion of the particle is included through the effective potential $U_B = k_B T \ln\psi$, written in terms of the distribution function of the particle, ψ. The orientation of the particle is represented using a right-handed, orthonormal set of three vectors $\mathbf{u}_1$, $\mathbf{u}_2$, and $\mathbf{u}_3$ fixed to the particle. The viscoelastic properties are calculated by assuming no external forces and that ψ is independent of the position of the particle.

Using this framework, the hydrodynamic stress per particle is

$$(S_H)_{ij} = K_{ijkl}\kappa_{kl} \tag{2}$$

and the Brownian stress per particle is

$$(S_B)_{ij} = h_{ijk}\mathcal{R}_k(-k_B T \ln\psi), \tag{3}$$

where K and h are mobility tensors and in the above equations and henceforth all indices used indicate the laboratory frame components, and repeated indices are summed unless otherwise stated. We also used the rotational derivative operator, $\boldsymbol{\mathcal{R}} = \mathbf{u}_\mu \times \partial/\partial\mathbf{u}_\mu$, where the index $\mu = 1, 2, 3$ is summed.

In general the stress tensor for the system is written as

$$\sigma = -p\mathbb{I} + 2\eta_s\kappa - n\langle S_B + S_H\rangle, \tag{4}$$

where p is the pressure, $\mathbb{I}$ the 3×3 identity matrix, and η_s the solvent viscosity. The angle brackets denote averaging over the orientational distribution $\langle\cdots\rangle = \int d\mathbf{u}_1 d\mathbf{u}_2\ \psi\ (\cdots)$. All viscoelastic properties follow from these expressions.

III. SYMMETRY CONDITIONS

A suspension of particles can be Newtonian if the Brownian and hydrodynamic stresses satisfy certain conditions. In the linear regime, the hydrodynamic stress contributes a constant to the complex viscosity and is therefore always a purely viscous, Newtonian contribution. The Brownian stress depends implicitly on the strain, via ψ, and can be shown to decay with time, contributing both real and imaginary parts to the complex viscosity. Therefore, in the linear regime, if the Brownian stress vanishes, the solution is purely viscous and Newtonian.

We consider the particles to be suspended in an incompressible fluid; therefore, the isotropic part of the fluid stress tensor is determined solely by the external conditions and is irrelevant for the viscoelasticity. As such, if the Brownian stress tensor becomes isotropic, then it may be taken to vanish without loss of generality, and the suspension is purely viscous.

For larger strains the hydrodynamic stress can produce non-Newtonian stresses. However, if the mobility tensor, K, is independent of the particle's orientation then its average must be independent of time and therefore $\langle S_H\rangle$ is purely viscous. For K to be independent of the particles' orientation in the laboratory frame it must be an isotropic tensor, so that it is invariant under all rotations from one orientation to another.

We therefore have two conditions which, if both are satisfied, are sufficient for the rheology to be Newtonian for all strain sizes: (I) the Brownian stress tensor must be isotropic and (II) the mobility tensor K must be isotropic. These statements are true assuming the particle distribution remains uniform and interparticle interactions are negligible. If we only require the suspension to be purely viscous in the linear regime, then only the first condition needs to hold.

To determine which particle shapes satisfy these conditions we consider the symmetry group, $\mathcal{G}$, of the particle. This is a set of 3×3 rotation matrices, R, which leave the orientation of the particle unchanged. For example, consider a cubic particle whose faces and edges are all identical and whose density is uniform. If we rotate this particle by $\pi/2$ about an axis passing through the center of any one of its faces, the orientation of the particle is outwardly the same. The only difference is the definition of the unit vectors, $\mathbf{u}_{1,2,3}$.

Under the action of R, the vectors' components transform according to the standard rule,

$$u_i^\mu \rightarrow (u')_i^\mu = R_{ai} u_a^\mu, \tag{5}$$

where the symbol u_i^μ denotes the ith component of the vector $\mathbf{u}_\mu$.

The physical consequence of these symmetries is that by applying the above transformation to the orientation vectors and imposing the same background fluid flow, the response of the particle would be the same as measured in the laboratory frame. Specifically, S_B and S_H are preserved under the action of the transformation (5).

Beginning with S_B, we use Eq. (3) and write it in terms of the transformed vectors $\mathbf{u}'$,

$$(S_B)_{ij} = -R_{ai} R_{bj} R_{ck} h_{abc} R_{dk} \mathcal{R}_d U_B, \tag{6}$$

where we have taken into account that $\mathcal{R} U_B$ and h transform as a psuedovector and third rank tensor, respectively, and $|R| = 1$.

We may then use the identity, $R_{dk} R_{ck} = \delta_{cd}$, to find that the Brownian stress must satisfy

$$(S_B)_{ij} = (\mathbf{R}^T \cdot \mathbf{S}_B \cdot \mathbf{R})_{ij}. \tag{7}$$

This must hold for each member of the shape's symmetry group; therefore, the following commutation relations are implied:

$$[\mathbf{S}_B, \mathbf{R}] = 0, \quad \forall\, \mathbf{R} \in \mathcal{G}. \tag{8}$$

We now make use of Schur's First Lemma [19], which states that if a matrix commutes with all members of an irreducible representation of a group, then it is proportional to the identity matrix. An irreducible representation is one where each member cannot be cast in block diagonal form by the same similarity transformation [20].

Condition (I) can now be understood as a condition on the particle shape; its symmetry group *must* have an irreducible representation in 3×3 matrices. If it does, the relations (8) and Schur's First Lemma imply $\mathbf{S}_B \propto \mathbb{I}$.

To ensure that a dilute suspension of particles is Newtonian for all strain sizes, condition (II) must be met. The relation between the hydrodynamic stress and the strain via K is mathematically the same as that between the stress, strain, and elastic modulus for solids, with K playing the role of the elastic modulus. The symmetry conditions needed for the solid elastic modulus to be isotropic have been widely studied [21,22]. By considering how the symmetries reduce the number of independent components of the tensor K, it has been shown that when K is invariant under a symmetry group with an irreducible representation of degree five it is isotropic [22]. This is true for spheres, icosahedra, and higher symmetry shapes, but not for cubes or lower symmetry shapes. Since shapes with icosahedral symmetry also satisfy condition (I), they will be purely viscous and Newtonian for all strain sizes, whereas shapes with cubic or tetrahedral symmetry will only have this property in the small strain regime.

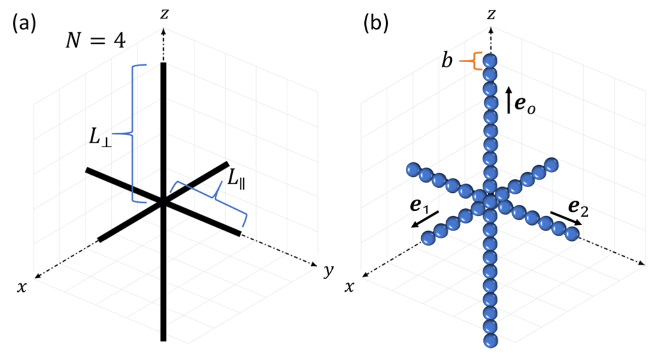

FIG. 2. (a) One of the wire-frame particles considered, with $N = 4$ in-plane legs and unequal lengths of the in and out of plane legs, $L_\parallel \neq L_\perp$. (b) The "shish kebab" procedure as applied to the shape from panel (a). The wire frame is replaced by spheres with diameter b placed along each leg of the shape. The unit vectors $\mathbf{e}_l$ associated with some of the legs are also indicated.

We can now understand why a dilute suspension of spheres is purely viscous, whereas rigid, rodlike particles have a viscoelastic linear response in dilute suspension. The symmetry group of a sphere, $O(3)$, has a well known irreducible representation in 3×3 matrices; hence the elasticity *must* vanish. Rods, on the other hand, are rotationally symmetric about their axis, chosen to align with $\mathbf{u}_3$, and are symmetric under the inversion, $\mathbf{u}_3 \rightarrow -\mathbf{u}_3$. These symmetries do not have an irreducible representation in 3×3 matrices. Therefore, dilute suspension of rods *can* have a finite viscoelasticity.

Figure 1(a) shows examples of shapes that produce purely viscous dilute suspensions for all strain sizes and small strains. Next, we construct a particular class of shapes not found by this symmetry argument whose dilute suspensions have a purely viscous stress response to small strains.

IV. WIRE FRAME PARTICLES

We consider shapes comprised of rigid, thin rods or legs. Each leg is indexed by l and has a different length L_l. We call these shapes "wire frames." We only consider shapes where legs meet at one point, see Fig. 2, but the formulas can be easily modified when this is not the case.

These particles provide a simple way to test our predictions and construct shapes of different symmetries within the same framework. They are also presented as a model for recently synthesized DNA nanoparticles [14]. Since double stranded DNA is very rigid with a persistence length of ∼390 Å [23] and a typical aspect ratio ∼20, the approximation of rigidity and large aspect ratios should be appropriate. We construct a formulation, based on Onsager's variational principle [15–18], to describe these shapes in general, into which any given shape may be specified.

The principle states that the linear and angular velocities of the particle are those which extremize the Rayleighian, $\mathcal{L}$, of the system. The Rayleighian is defined as $\mathcal{L} = \dot{F} + \frac{1}{2}\Phi$, where $\dot{F}$ is the time derivative of the Helmholz free energy and Φ is the energy dissipation function.

To determine the Rayleighian we use a standard "shish kebab" approach [3]. We place N_l spherical beads of radius b

along each leg of the shape, such that they just touch, i.e., the length of the lth leg is $L_l = N_l b$. Figure 2(b) shows this for a particular wire-frame shape. The position vector and velocity of the nth bead on leg l are written as

$$\mathbf{r}_n^l = \mathbf{r} + nb\,\mathbf{e}_l, \quad \mathbf{v}_n^l = \mathbf{v} + nb\,\mathbf{\Omega} \times \mathbf{e}_l, \tag{9}$$

where $n \in [0, N_l]$ and the unit vector $\mathbf{e}_l$ points along the leg.

The velocities of all the beads are in general coupled together by hydrodynamic interactions. This coupling is difficult to handle exactly, but a simple approximation can be made. We assume that the rods are very long compared to their width, such that the majority of beads on different legs are very far apart. This allows the hydrodynamic interaction between beads on different legs to be neglected, which becomes an accurate approximation in the limit of infinite aspect ratio, L_l/b.

Using a textbook procedure [3,18] the Rayleighian for a general wire-frame shape can be determined, providing a complete description of its behavior (see Supplemental Material [24]). We focus on the linear viscoelastic properties, so we only consider terms containing the angular velocity. The relevant terms in the Rayleighian are

$$\mathcal{L} = \langle \mathbf{\Omega} \cdot \boldsymbol{\mathcal{R}} k_B T \ln\psi \rangle + \frac{1}{2}\left\langle \sum_l \lambda_l (\mathbf{\Omega} \times \mathbf{e}_l) \cdot (\mathbf{v} - \kappa \cdot \mathbf{r}) \right\rangle + \frac{1}{2}\left\langle \sum_l \mu_l\, (\mathbf{\Omega} \times \mathbf{e}_l)^2 - 2\mu_l\, \mathbf{\Omega} \cdot (\mathbf{e}_l \times \kappa \cdot \mathbf{e}_l) \right\rangle + (\cdots), \tag{10}$$

where we have defined the friction constants λ_l and μ_l as

$$\lambda_l = \frac{4\pi \eta_s L_l^2}{\ln(L_l/b)}, \quad \mu_l = \frac{8\pi \eta_s L_l^3}{3\ln(L_l/b)}. \tag{11}$$

By appropriately choosing the unit vectors, $\mathbf{e}_l$, we can describe any wire-frame shape.

We specifically consider shapes comprised of N evenly spaced, coplanar legs of equal length with two antiparallel legs pointing orthogonally out of plane, as shown in Fig. 2. The lengths of the in and out of plane legs are $L_\parallel$ and $L_\perp$, respectively. The in plane legs are separated by an angle $\phi = 2\pi/N$. The unit vectors parallel to the legs in plane are denoted $\mathbf{e}_l = \cos(l\phi)\mathbf{u}_1 - \sin(l\phi)\mathbf{u}_l$ for $1 \leqslant l \leqslant N$. For the out of plane legs, $\mathbf{e}_{-1,0} = \pm\mathbf{u}_3$.

According to Onsager's principle [15–18], the components of the stress tensor are given by $\sigma_{ij} = \partial\mathcal{L}^*/\partial\kappa_{ij}$, where $\mathcal{L}^*$ is the Rayleighian (10) evaluated at the extremum value of $\mathbf{\Omega}$. We find that the Brownian stress tensor as a function of time is given by (see Supplemental Material [24])

$$(S_B)_{ij} = nk_B T\, G \int_{-\infty}^{t} dt'\, e^{-(t-t')/\tau} \kappa_{ij}(t'), \tag{12}$$

where τ is a decay timescale and G is the elastic modulus

$$G = 6\left(\frac{N - 4\gamma}{N + 4\gamma}\right)^2, \tag{13}$$

with $\gamma \equiv \mu_\perp/\mu_\parallel$. The friction constants, $\mu_\parallel$ and $\mu_\perp$, are calculated from (11) for the in- and out-of-plane legs, respectively. This modulus is plotted in Fig. 3(b) for $N = 4$ and $N = 12$.

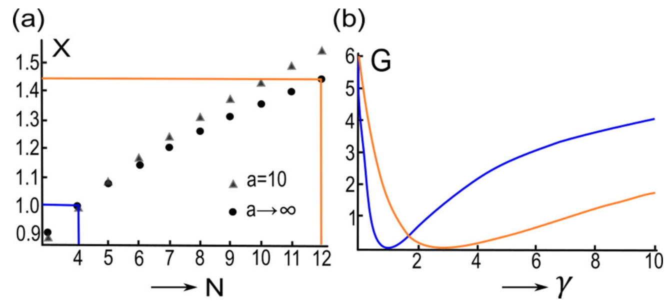

FIG. 3. (a) Required ratio of leg lengths, $x = L_\perp/L_\parallel$, for the elasticity to vanish plotted as a function of the number of in-plane legs, $3 \leqslant N \leqslant 12$, for aspect ratios, $a = L_\perp/b = 10$ (triangles) and $a \to \infty$ (circles). These ratios are solutions to (14) for x, which in the case $a \to \infty$ are simply $x = (N/4)^{1/3}$. The (dark) blue and (light) orange lines indicate the solutions for $N = 4$ and $N = 12$, respectively. Panel (b) shows the elastic modulus, G, as given in (13), plotted as a function of $\gamma \approx L_\perp^3/L_\parallel^3$. The blue curve corresponds to $N = 4$ and the orange $N = 12$.

The elastic modulus has a minimum of zero when $\gamma = N/4$. This means that when the ratio of leg lengths, $x = L_\perp/L_\parallel$, satisfies the transcendental equation,

$$4x^3 \ln(a/x) = N \ln a, \tag{14}$$

the suspension is purely viscous. The aspect ratio a is defined as $L_\perp/b$. This recovers the expected result; a dilute suspension of symmetric cross shapes with $N = 4$ and $L_\perp = L_\parallel$ (equivalent to an octahedron or a cube) has a purely viscous linear stress response.

A plot of the solutions to this equation for $3 \leqslant N \leqslant 12$ and $a = 10$ is shown in Fig. 3(a) (triangles); we also show the solutions in the limit $a \to \infty$ (circles), which allows γ to be approximated as x^3.

It is intriguing that we can engineer the elasticity to vanish for *any* N by choosing the right ratio, x. For instance, when $N = 3$ the particle has the symmetry of a trigonal bipyramid of variable height. The symmetry group for such an object does not satisfy the conditions given previously, yet when the ratio of lengths is chosen appropriately the elasticity still vanishes, in the small κ regime.

This phenomenon, while not explained by a simple symmetry argument, can be physically understood by considering the stresslet produced by the rotation of the particle. When a rod rotates about an axis perpendicular to its length, the surrounding fluid flows towards the ends, but away from the broad side of the rod [3]. The resulting flow is typical of the stresslet singularity, whose magnitude depends on the length of the rod.

When a planar cross rotates, the stresslet flows cancel each other in the plane of the shape because the two equal length rods perpendicularly bisect each other. This construction may be applied to the $N = 4$, $\gamma = 1$ wire frame. Shapes where $N \neq 4$ can be engineered to produce no stresslet by tuning the rod lengths such that the out of plane rod produces a stresslet which cancels that produced by those in plane.

V. CONCLUSIONS

We have discussed the origins of non-Newtonian rheology for dilute suspensions of rigid particles and determined sufficient conditions the shape must satisfy for a dilute suspension to be Newtonian. To have a purely Newtonian response for all strain sizes the symmetry group of the shape must have irreducible representations of degree 3 and degree 5, whereas in the regime of small strains the symmetry group only needs an irreducible representation of degree 3 for Newtonian behavior. This allows for simple classification of suspensions without the need for detailed calculation.

We also developed a framework for studying the rheology of wire-frame particles constructed from thin, rigidly connected rods using Onsager's variational principle. This was used to demonstrate the vanishing elasticity for octahedral and cubic shapes in the linear regime, as well as find a set of bipyramidal shapes which, despite their symmetry group not satisfying the appropriate condition, have Newtonian dilute suspensions for small strains. This is physically explained in terms of the stresslets produced by the rotation of each constituent rod.

The study of wire-frame shapes has relevance for the design of DNA nanostar suspensions, where understanding which particle shapes lead to a particular rheological response is very important.

ACKNOWLEDGMENTS

We are grateful to Prof. D. Frenkel for insightful conversations and valuable comments on the manuscript. D.A.K. acknowledges financial support from the UK Engineering and Physical Sciences Research Council Ph.D. studentship Award No. 1948692.

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