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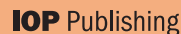
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Rheological properties of a dilute suspension of self-propelled particles

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Abstract – With a detailed microscopic model for a self-propelled swimmer, we derive the rheological properties of a dilute suspension of such particles at small Peclet number. It is shown that, in addition to Einstein's like contribution to the effective viscosity, that is proportional to the volume fraction of the swimmers, a contribution due to the activity of self-propelled particles influences the viscosity. As a result of the activity of swimmers, the effective viscosity would be lower (higher) than the viscosity of the suspending medium when the particles are pusher (puller). Such activity-dependent contribution will also result a non-Newtonian behavior of the suspension in the form of normal stress differences.

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Introduction. – Dynamical properties of bacterial suspensions and suspensions of artificially designed self-propelled micro-particles have been the subject of many recent experimental and theoretical investigations [1–4]. In addition to the self-organized behavior which have been observed in such active suspensions [5–12], it is an essential task to understand how such active suspensions respond to external forces and what rheological behavior they have [13–15]. Rheology of active biological matter composed of self-propelled particles is important and interesting from a fundamental point of view, as in such systems the particles inject mechanical energy into the ambient fluid without applying any net hydrodynamical forces. Understanding the physics behind such phenomena is important for micro-fluidic experiments that manipulate samples of microorganisms and also it could be relevant for micro-robots that are artificially developed.

Among all macroscopic rheological parameters of the system, the effective viscosity of such complex fluids is the main core of current investigations [16,17]. It is a known experimental fact that the effective viscosity shows different behavior for active suspensions containing swimmers whose motion is generated by head (puller) or tail (pusher). A recent experiment on suspension of motile algae *Clomydomonas* shows that puller particles increase the effective viscosity [18]. The effects of pusher particles are

examined in an experiment performed on bacterium *Bacillus subtilis* [19]. It is shown that at small volume fraction of swimmers, the effective viscosity is smaller than the viscosity of the ambient fluid, but at large volume fraction, the viscosity would be larger than the bare viscosity. In another experiment on *E. coli*, it is shown that at small Peclet number, the effective viscosity of pushers is smaller than the bare viscosity of the fluid [20].

Most of the theoretical and numerical works which have been done so far are theories with phenomenological origin [14,16,21,22]. In this letter, we use a microscopic model for pusher and puller particles and investigate their influence on the rheology of the suspension. This kind of description will allow us to have a detailed insight into the role of the microscopic parameters of the swimmer in the rheological properties.

To study the rheological properties of a dilute suspension of microswimmers, we first study the dynamics of a single swimmer suspended in a Newtonian fluid with viscosity η_0 that is subject to an external shear. This will allow us to eventually achieve the response of the fluid to the external forces. As a mathematical model, a three-sphere swimmer can model the general hydrodynamical characteristics of most low Reynolds swimmers [23,24] and it captures the peculiarities at low Reynolds hydrodynamics [25–27]. Figure 1 shows a schematic view of a three-sphere swimmer immersed in a purely straining shear flow. The swimmer consists of three spheres with

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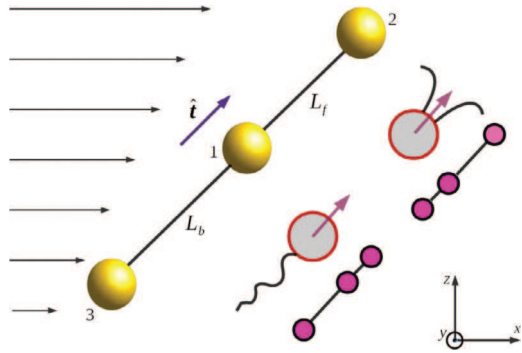


Fig. 1: (Colour on-line) Geometry of a low Reynolds swimmer immersed in a shear flow. For an asymmetric swimmer whose back and front arms have different sizes, the three-sphere swimmer can capture the hydrodynamics of both pusher and puller active particles. A puller, e.g., *Clomydomonas*, corresponds to $\langle L_f - L_b \rangle_t > 0$, and a pusher, e.g., *Bacillus subtilis*, corresponds to $\langle L_f - L_b \rangle_t < 0$.

radii a linked by two front and back arms with variable lengths $L_f(t) = L + u_1(t)$ and $L_b(t) = (1 + \delta)L + u_2(t)$. Here L and $(1 + \delta)L$, denote the mean arm lengths and $u_1(t)$ and $u_2(t)$ are two periodic functions of time with same amplitudes u_0 . We further assume that these periodic functions average to zero. By considering an asymmetry parameter δ , we will show how the asymmetry of the swimmer is essential in the overall dynamics of the system. The instantaneous orientation of the swimmer is denoted by a unit vector $\hat{\mathbf{t}}$ that points from the swimmer's back to its front side. In terms of polar variables, this vector can be written as $\hat{\mathbf{t}} = (\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta)$, where θ and φ are the polar variables. The externally applied shear flow is given by $\mathbf{u}^s(\mathbf{r}) = \Gamma \cdot \mathbf{r}$, with $\Gamma = \dot{\gamma} \hat{\mathbf{z}} \hat{\mathbf{x}}$, where $\dot{\gamma}$ represents the shear rate. In the presence of the swimmer, the velocity field of the fluid will change. To express the velocity field in the presence of the swimmer, we consider that the sphere's diameter a is much less than the arm's lengths. Denoting the force exerted on fluid from the j -th sphere that is located at position \mathbf{r}_j by \mathbf{f}_j , the velocity field at a given point \mathbf{r} reads as

$$\mathbf{u}(\mathbf{r}) = \sum_{j=1}^3 \mathcal{O}(\mathbf{r} - \mathbf{r}_j) \cdot \mathbf{f}_j + \Gamma \cdot \mathbf{r} + \mathbf{C}(\mathbf{r}) : \Gamma, \quad (1)$$

where $\mathcal{O}(\mathbf{r}) = (1/8\pi\eta_0 r)(\mathbf{I} + \hat{\mathbf{r}}\hat{\mathbf{r}})$ is the Green's function of the Stokes equation [28]. Note that the Green's function regularizes to $\mathcal{O}(0) = (1/6\pi\eta_0 a)\mathbf{I}$. The third-rank disturbance tensor \mathbf{C} represents the hydrodynamic interaction of two spheres in a shear flow and for sphere i this disturbance matrix is given by [29]

$$\mathbf{C}(\mathbf{r}_i) = \sum_{j \neq i} \left(-\frac{5}{2} \frac{a^3}{|\mathbf{r}_{ij}|^3} + \frac{20}{3} \frac{a^5}{|\mathbf{r}_{ij}|^5} \right) \frac{\mathbf{r}_{ij} \mathbf{r}_{ij} \mathbf{r}_{ij}}{|\mathbf{r}_{ij}|^2} - \frac{4}{3} \frac{a^5}{|\mathbf{r}_{ij}|^5} \left[\mathbf{I} \mathbf{r}_{ij} + (\mathbf{I} \mathbf{r}_{ij})^\dagger \right] - \frac{25}{2} \frac{a^6}{|\mathbf{r}_{ij}|^6} \frac{\mathbf{r}_{ij} \mathbf{r}_{ij} \mathbf{r}_{ij}}{|\mathbf{r}_{ij}|^2}, \quad (2)$$

where $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$, and $\hat{\mathbf{r}}_{ij} = (\mathbf{r}_i - \mathbf{r}_j)/|\mathbf{r}_i - \mathbf{r}_j|$. We are seeking the autonomous net swimming motion of the system. Denoting the velocity of the i -th sphere by \mathbf{v}_i , the no-slip boundary condition on the spheres requires that $\mathbf{u}(\mathbf{r}_i) = \mathbf{v}_i$. The internal dynamics of the arms are imposed by the following geometrical constraints:

$$\mathbf{r}_2 - \mathbf{r}_1 = L_f(t) \hat{\mathbf{t}}, \quad \mathbf{r}_1 - \mathbf{r}_3 = L_b(t) \hat{\mathbf{t}}. \quad (3)$$

As there is no externally applied force or torque on the swimmer, the system should be force and torque free in the sense that $\mathbf{f}_1 + \mathbf{f}_2 + \mathbf{f}_3 = 0$ and $\mathbf{r}_{21} \times \mathbf{f}_2 + \mathbf{r}_{31} \times \mathbf{f}_3 = 0$. Now we can eliminate the dynamics of the background fluid and study the dynamics of the swimmer. We make a further mathematical simplification by assuming that the changes in the arm lengths are small: $L_i \gg u_1, u_2, a$ ($i = f, b$). Throughout what follows, we will keep only the leading-order terms in terms of these small quantities. One note that this Taylor expansion is performed to simplify the mathematical analytical results and it does not change the generic features of the results. We can express the overall dynamics in terms of the internal motions given by $u_1(t)$ and $u_2(t)$. The time-averaged linear velocity of the swimmer's center up to the leading order of the small quantities u_1/L , u_2/L and a/L reads

$$\langle \mathbf{V}_s \rangle_t = \dot{\gamma} z \hat{\mathbf{i}} + (V^0 + V^\dot{\gamma}) \hat{\mathbf{t}}. \quad (4)$$

Here, $V^0 = -\frac{7}{24} \frac{a}{L^2} (1 - \delta) \Phi$ is the swimming speed of a swimmer in a quiescent flow, and $V^\dot{\gamma} = -\frac{7}{4} a \dot{\gamma} \lambda \delta$ represents the velocity changes due to the presence of a shear flow. Here $\Phi = \langle u_1 \dot{u}_2 - u_2 \dot{u}_1 \rangle_t$ and $\lambda = \sin \theta \cos \theta \cos \varphi$. We have further assumed that the asymmetry parameter δ is also very small. Externally applied shear flow, tends to rotate the swimmer. The angular velocity of the swimmer in polar coordinates can be written as $\vec{\omega} = \dot{\theta} \hat{\boldsymbol{\theta}} + \dot{\varphi} \sin \theta \hat{\boldsymbol{\varphi}}$, with

$$\dot{\theta} = \dot{\gamma} \cos^2 \theta \cos \varphi, \quad \dot{\varphi} = -\dot{\gamma} \cot \theta \sin \varphi. \quad (5)$$

This is the same result which is obtained for the rotational velocity of a slender body immersed in an external shear flow, namely the well-known Jeffery orbits [30]. In addition to the swimming velocity, we can calculate the hydrodynamic forces exerted on the fluid by the spheres. To express the forces, we decompose all forces as $\mathbf{f}_i = \mathbf{f}_i^0 + \mathbf{f}_i^\dot{\gamma}$ where the contribution due to the shear flow is explicitly separated from the terms that are present for a swimmer moving in a quiescent flow. Time averaged forces for a swimmer moving in a quiescent flow are given by

$$\langle \mathbf{f}_2^0 \rangle_t = -\frac{5}{8} \pi \eta_0 \left(\frac{a}{L} \right)^2 \left(1 + \frac{7}{5} \delta \right) \Phi \hat{\mathbf{t}},$$

$$\langle \mathbf{f}_3^0 \rangle_t = -\frac{5}{8} \pi \eta_0 \left(\frac{a}{L} \right)^2 \left(1 - \frac{17}{5} \delta \right) \Phi \hat{\mathbf{t}},$$

and the contributions due to the shear flow are given by

$$\langle \mathbf{f}_2^\dot{\gamma} \rangle_t = -6\pi\eta_0 a L \lambda \dot{\gamma} \left(1 + \frac{1}{3} \delta \right) \hat{\mathbf{t}},$$

$$\langle \mathbf{f}_3^\dot{\gamma} \rangle_t = 6\pi\eta_0 a L \lambda \dot{\gamma} \left(1 + \frac{2}{3} \delta \right) \hat{\mathbf{t}}.$$

We will use the above results to extract the rheological response of a fluid containing a collection of such swimmers. But before studying the properties of a suspension, we note that the force-dipole tensor for a swimmer moving in a quiescent flow can be written as $\mathbf{D} = 6\pi\eta_0(a/L)^2\delta L\Phi\hat{\mathbf{t}}\hat{\mathbf{t}}$. It is shown that the far-field velocity of a swimmer can be expanded in terms of the moments of the force [31]. As one can see for an asymmetric swimmer with $\delta \neq 0$, the force dipole velocity field dominates the far-field behavior. An intuitive classification of the swimmers is based on the observation of how the driving force of the motion is located at the head or at the tail of the swimmer. For pushers, the driving force sits on the tail while for pullers, the driving force sits on the head. Referring to fig. 1, for $\Phi < 0$, the intrinsic velocity of the swimmer is in the direction given by $\hat{\mathbf{t}}$, and it is the sign of δ which determines whether the swimmer is puller or pusher. For $\delta > 0$ (< 0) the swimmer is pusher (puller).

After calculating the forces exerted by the spheres to the fluid, the contribution to the stress tensor of the fluid from a single swimmer can be obtained. For a dilute suspension of N objects (swimmers) moving in an ambient fluid with viscosity η_0 , the stress tensor averaged over the surface of objects can be written as $\sigma = \sigma^0 + \frac{N}{V}\mathcal{S}$ [32], where σ^0 represents the stress tensor if all the objects are removed from the fluid. The tensor \mathcal{S} is the extra stress due to the presence of a single object and it reads

$$\mathcal{S} = - \int \left[\frac{1}{2}(\mathbf{r}\boldsymbol{\sigma} \cdot \hat{\mathbf{n}} + \boldsymbol{\sigma} \cdot \hat{\mathbf{n}}\mathbf{r}) - \frac{1}{3}(\mathbf{r} \cdot \boldsymbol{\sigma} \cdot \hat{\mathbf{n}})\mathbf{I} \right] d\mathbf{A},$$

where the integral is over the surface of the body and $\boldsymbol{\sigma} \cdot \hat{\mathbf{n}}$ is the stress at any point \mathbf{r} on the body. In the present case, each object is a collection of three connected spheres and in the limit of approximations performed before, they are replaced by point forces. In terms of the forces exerted by spheres, this stress tensor reads $\mathcal{S} = -\mathbf{r}_{21}\mathbf{f}_2 - \mathbf{r}_{31}\mathbf{f}_3$. Inserting the results given before and averaging over time we can keep the leading-order terms:

$$\langle \mathcal{S} \rangle_t = -\frac{29}{4}\pi\eta_0 \frac{a^2}{L} \Phi \delta \hat{\mathbf{t}}\hat{\mathbf{t}} + 12\pi\eta_0 aL^2 \dot{\gamma} (1 + \delta) \lambda \hat{\mathbf{t}}\hat{\mathbf{t}}.$$

The first part in the above stress is independent of the shear rate ($\dot{\gamma}$). We call this part as active contribution to the stress tensor, in the sense that it depends on the internal activity of the swimmer. The second part of the stress tensor reflects a shear-rate-dependent contribution that is a passive part. There is no cross-term in the stress tensor with simultaneous dependence both on the swimmer's activity and the shear rate. This is due to the linearity of the Stokes equation that does not allow any direct combination between the external stress and the internal activity of the swimmer. As one can distinguish, the stress contribution from a swimmer depends crucially on the angular direction of the swimmer. Assuming that our system contains a collection of randomly orientated swimmers, we can average the stress tensor over all swimmers to reach

the following result for the off-diagonal elements of the total stress tensor:

$$\sigma_{ij} = \frac{\eta^{\text{eff}}}{2} \dot{\gamma} (\delta_{i,x}\delta_{j,z} + \delta_{j,x}\delta_{i,z}).$$

Comparing this result with the corresponding components of the stress tensor for a simple shear flow, we can easily conclude that a dilute suspension of swimmers behaves like a Newtonian fluid where its viscosity is replaced by η^{eff} . The effective viscosity of a homogeneous and dilute suspension of passive swimmers is obtained as

$$\eta^{\text{eff}} = \eta_0 \left(1 + \frac{5}{2}c \left(\frac{L}{5a} \right)^2 \right),$$

where $c = 4\pi a^3 \frac{N}{V}$ is the volume fraction of N swimmers distributed in a fluid with volume V . This result is comparable with Einstein's relation for the effective viscosity of a dilute suspension of objects [32]. The original Einstein's relation is valid for spherically symmetric objects. Here the term $(\frac{L}{5a})^2$ shows that a single swimmer is not spherically symmetric. However if we want to think about an effective volume fraction for the swimmers, because of the elongational asymmetry, the effective volume fraction will be much larger than the real volume fraction of isolated swimmers that are composed of three spheres. Such a similar result for the effective viscosity of a dilute suspension of asymmetric particles was observed for ellipsoidal particles [33]. It should be noted that the contribution from the swimmer's activity (the active part in the stress tensor) averages to zero and it does not have any influence on the effective viscosity of the suspension. This is understandable in the sense that the viscosity measures the linear response of the fluid to an externally applied shear, and the active term in the stress does not depend on the shear rate at all. The appearance of any cross-term in the stress tensor would cause the suspension to have an effective viscosity proportional to the activity of swimmers. But here, linearity of the Stokes equation does not allow any such contribution. Please note that this point of view is in contrast with the phenomenology that is used in previous works [34].

Our central question in this work is the influence of the swimmers activity on the fluid rheological properties. Here we will show that, taking into account the thermal fluctuations of the swimmers, we will introduce a mechanism that can couple the internal activity of the swimmers to the externally applied shear rate. This will result in a non-trivial non-Newtonian behavior for the fluid. To study the thermal fluctuations of the swimmers, we denote the angular distributions of such swimmers by $F(\theta, \varphi)$. In addition to time averaging over the internal motion of an individual swimmer, we need to average over the angular distribution as well. Having in hand the distribution function, we can perform the averaging procedure for a function $\delta\mathcal{S}$ as follows:

$$\langle \delta\mathcal{S} \rangle_{t,e} = \left\langle \int d\Omega F(\theta, \varphi) \delta\mathcal{S} \right\rangle_t, \quad (6)$$

where the integral should be taken over the polar angles. The distribution function obeys the following Fokker-Planck equation:

$$\frac{\partial F}{\partial t} = D_r \nabla^2 F - \nabla \cdot (F \vec{\omega}), \quad (7)$$

where ∇^2 is the angular part of the Laplacian operator and the velocity in the angular space is given by $\vec{\omega} = \dot{\theta} \hat{\theta} + \dot{\varphi} \sin \theta \hat{\varphi}$. Here D_r (in s^{-1}) stands for the rotational diffusion coefficient of the swimmers and it depends both on the thermal energy $k_B T$ and on the rotational friction coefficient of the swimmer ξ_r by $D_r = k_B T / \xi_r$. The rotational friction coefficient is a hydrodynamical quantity that measures the response of the swimmer to an externally applied torque. Direct calculations for a three-sphere system immersed in a quiescent fluid and subject to an external torque, reveals that $\xi_r = 4\pi\eta_0 a(L_b^2 + L_f^2 + L_b L_f)$. In terms of small dimensionless Peclet number $Pe := (\dot{\gamma}/k_B T)\xi_r$, we can set up a perturbation steady-state solution for the above Fokker-Planck equation as

$$F(\theta, \varphi) = \frac{1}{4\pi} \left[1 + \frac{Pe}{2} (\sin 2\theta \cos \varphi) + \frac{Pe^2}{280} \left((7 - 35 \cos^4 \theta) + (35 \sin^2 \theta \cos^2 \theta + 5 \sin^2 \theta) \cos 2\varphi \right) + \dots \right]. \quad (8)$$

The Peclet number is essentially the ratio between the diffusion time $\tau_D = D_r^{-1}$ and the time scale of the applied shear flow $\tau_s = \dot{\gamma}^{-1}$. Using the above distribution function, we can calculate all statistical variables up to the second order of small parameter Pe . To obtain the rheological properties of the suspension, we should repeat calculations similar to what we have done before eq. (6). Along such calculations we should perform an ensemble averaging followed by the time average over the internal configuration. We will need the following ensemble averages:

$$\langle \hat{\mathbf{t}} \hat{\mathbf{t}} \rangle_{t,e} = \begin{pmatrix} \frac{1}{3} + \frac{1}{105} Pe^2 & 0 & \frac{1}{15} Pe \\ 0 & -\frac{1}{120} Pe^2 & 0 \\ \frac{1}{15} Pe & 0 & \frac{1}{3} - \frac{23}{840} Pe^2 \end{pmatrix},$$

and

$$\langle \lambda \hat{\mathbf{t}} \hat{\mathbf{t}} \rangle_{t,e} = \begin{pmatrix} \frac{1}{35} Pe & 0 & \frac{1}{15} + \frac{52}{315} Pe^2 \\ 0 & \frac{1}{105} Pe & 0 \\ \frac{1}{15} + \frac{52}{315} Pe^2 & 0 & \frac{1}{35} Pe \end{pmatrix}.$$

Using the above results, it is straightforward to calculate the rheological properties of the dilute suspension of swimmers. The effective viscosity of the suspension is the first and most important quantity that we can address here. For a dilute suspension of swimmers with volume fraction

c , that was defined before, the effective viscosity reads

$$\eta^{\text{eff}} = \eta_0 \left[1 + c \left(\frac{5}{2} \left(\frac{L}{5a} \right)^2 - \frac{W}{k_B T} \delta \left(\frac{L}{a} \right) \right) \right], \quad (9)$$

where $W = \frac{213\pi}{14} \eta_0 L^2 \bar{v}$, and $\bar{v} = -\frac{7}{24} \frac{a}{L^2} \Phi$. At a very high value of temperature, where the effects of thermal fluctuations dominate, we can recover the previous Einstein's like behavior for the viscosity of suspension. The main result that we obtain here is the influence of the swimmer's activity on the viscosity. The activity of the swimmer contributes to the viscosity through the function W . The asymmetry parameter δ appears in the above result, this is another important feature of our results which reflects the fact that the viscosity for pushers or pullers is different. For pushers (pullers), $\delta > 0$ ($\delta < 0$) and one can easily see that $\eta^{\text{eff}} < \eta^*$ ($\eta^{\text{eff}} > \eta^*$), where η^* is the viscosity of the suspending fluid enhanced by the passive contribution from swimmers. Such viscosity reduction and enhancement for a suspension of active particles are in clear agreement with the experiments that have been performed recently in the limit of small volume fraction [18–20,35]. In addition to the effective viscosity of the suspension, the fluctuation of the active particles will mediate an asymmetric behavior for the fluid. Normal stress differences quantify this non-Newtonian behavior of the suspension. Normal stress differences, up to second order in Peclet number, read

$$\begin{aligned} N_1 &= \sigma_{xx} - \sigma_{yy} = -W' \left(\frac{1}{3} + \frac{1}{56} \mathcal{P}^2 \right) - \frac{8}{35} (\pi \eta_0 a L^2 \dot{\gamma}) \mathcal{P}, \\ N_2 &= \sigma_{xx} - \sigma_{zz} = \frac{31}{840} W' \mathcal{P}^2, \end{aligned} \quad (10)$$

where $W' = (a/L)\delta W$. Note that these normal stress differences have different signs for pushers and pullers. This may reflect in standard rheological experiments, for example in the Weissenberg effect.

We should emphasize that thermal fluctuations and elongated geometry of the swimmer have crucial importance in the underlying physics presented here. Both these phenomena have been considered in phenomenological descriptions which are based on symmetry considerations [14]. The resident time of an elongated object in the preferred direction of the shear flow will be modified by the fluctuations and then, as predicted by phenomenological models, the effective viscosity changes. Here, by taking into account microscopic quadrupolar details for the swimmers, we obtained the results. The microscopic derivation of the results will allow us to have insights into the effects and roles of internal dynamical and geometrical parameters of the swimmer in the macroscopic rheological properties of the suspension.

To have an intuition about the mathematical results that we have obtained so far, we can think about the numerical values of the parameters. The diffusion time scale τ_D , the time scale for shear flow τ_s , and the internal time

scale for an individual swimmer given by $\tau_i = u_0^2/|\Phi|$, are three different time scales that characterize our system. τ_D is a passive parameter that crucially depends on the size of the swimmer. For a swimmer with the largest length $L \sim 1 \mu\text{m}$ and the smallest length $a \sim 0.1 \mu\text{m}$, we see that $\tau_D \sim 1 \text{ s}$. For a real microswimmer, for example spermatozoa, the undulation frequency of flagellum that is about 10 Hz, sets the internal time scale that is greater than the diffusion time. How important is the activity contribution to the effective viscosity? To answer this question, one can see from eq. (9), that the ratio between the active and passive correction to the viscosity is given by $\delta(a/L)(W/k_B T)$. Assuming that $\delta \sim a/L \sim 0.1$ and at room temperature, for a typical swimmer with swimming velocity $\bar{v} \sim 1 \mu\text{m/s}$, the above ratio is of the same order as the passive part.

To summarize, we have shown how a collection of thermally fluctuating swimmers can mediate an effective viscosity for the suspension. Depending on the type of individual swimmers, either pushers or pullers, the effective viscosity would be smaller or larger than the viscosity of the ambient fluid. In addition to the effective viscosity, the normal stress differences are also calculated for such suspension.

A natural extension of our work is the generalization to the systems with larger volume fractions. Active and passive hydrodynamic interactions between swimmers [36–38] may influence the viscosity for systems that are beyond the dilute regime considered here. One may imagine that in a suspension of microorganisms, another mechanism related to the internal feedback of the organism may provide an additional source of viscosity change. If we assume that an organism can change its internal undulation proportional to the rate of external shear, this scenario also will mediate an effective contribution to the viscosity.

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