Rheology of Active-Particle Suspensions

Yashodhan Hatwalne,[1] Sriram Ramaswamy,[2] Madan Rao,[1,3] and R. Aditi Simha[2,4]
[1]*Raman Research Institute, C.V. Raman Avenue, Bangalore 560 080, India*
[2]*Centre for Condensed Matter Theory, Department of Physics, Indian Institute of Science, Bangalore 560 012, India*
[3]*National Centre for Biological Sciences, UAS-GKVK Campus, Bellary Road, Bangalore 560 065, India*
[4]*MPI-PKS, Nöthnitzer Strasse 38, 01187 Dresden, Germany*
(Received 25 August 2003; published 19 March 2004)

We study the interplay of activity, order, and flow through a set of coarse-grained equations governing the hydrodynamic velocity, concentration, and stress fields in a suspension of active, energy-dissipating particles. We make several predictions for the rheology of such systems, which can be tested on bacterial suspensions, cell extracts with motors and filaments, or artificial machines in a fluid. The phenomena of cytoplasmic streaming, elastotaxis, and active mechanosensing find natural explanations within our model.

DOI: 10.1103/PhysRevLett.92.118101 PACS numbers: 87.16.Ac, 87.10.+e, 87.15.Ya

An active particle [1,2] absorbs energy from its surroundings or from an internal fuel tank and dissipates it in the process of carrying out internal movements usually resulting in translatory or rotary motion. This broad definition includes macroscopic machines and organisms, living cells, and their components such as actin-myosin and ion pumps [3]. In this Letter, we consider the interplay of activity, order, and flow via coarse-grained equations governing the hydrodynamic velocity, concentration, and stress fields in a suspension containing active particles of linear size ℓ, at concentration ϕ, each particle exerting a typical force f on the ambient fluid, with the activity of an individual particle correlated over a time τ_0 (the "run" time of a bacterium), and collective fluctuations in the activity correlated over length scales ξ and time scales τ. Rather than focusing on ordered phases [4], instabilities [4,5], or patterns (asters, vortices, spirals) formed in such assemblies [6] which our equations are of course capable of predicting, we apply them in the isotropic phase, with a view to understanding how a system such as a biological cell, composed of *active* elements, responds to deformation or mechanical stress. In addition to throwing light on full-cell rheometry [7,8], our equations form the framework for an analysis of any experiment probing the mechanical consequences of biological activity.

Our simple model makes rather interesting predictions: An orientationally ordered state [9] of active particles has a nonzero, macroscopic, anisotropic stress in contrast with thermal equilibrium nematics. Activity contributes an amount $\delta\eta \sim f\ell\phi\tau$ to the viscosity, with a sign determined by the type of active particle, and always enhances the apparent (noise) temperature. The latter greatly enhances the amplitude of the $t^{-d/2}$ long time tails [10] in the velocity autocorrelation. On approaching an *orientationally* ordered state, active suspensions with $\delta\eta > 0$ behave similar to passive systems near *translational* freezing, showing strong shear thickening and Maxwell-like viscoelasticity. Nonlinear fluctuation corrections give a dynamic modulus $G^*(\omega) \sim \sqrt{i\omega}$ for $\omega \gg \tau^{-1}$ observable over a large dynamic range, since τ is large. Cytoplasmic streaming [11], in which material flows from the depolymerizing trailing edge to the polymerizing leading edge of a crawling amoeboid cell, finds a natural explanation in our model, as do elastotaxis [12] and active mechanosensing [13], where cells orient their motion along preferred axes of the ambient medium.

These results follow from equations of motion based simply on the conservation law $\partial_t \mathbf{g} = -\boldsymbol{\nabla}\cdot\boldsymbol{\sigma}$ for the total (particles + fluid) momentum density $\mathbf{g}(\mathbf{x}, t)$ for an incompressible suspension. The stress tensor $\boldsymbol{\sigma}$ must in turn be determined by constitutive relations which emerge from an additional equation of motion for an active order-parameter field. We ignore, for simplicity, the dynamics of the active-particle concentration $\phi(\mathbf{x}, t)$, energy density, and nutrient fields.

To determine the contributions of activity to the stress, we need the forces associated with the active particles. Since there are no *external* forces on the system, the simplest active particle, on long time scales, is a permanent force dipole [14] (see Fig. 1). A collection of such

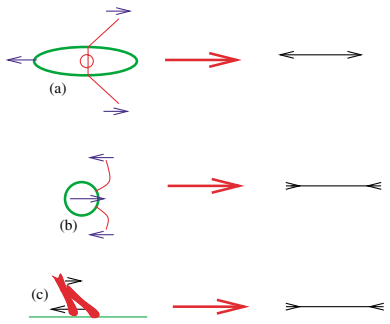

FIG. 1 (color online). Force dipoles for (a) a rowboat, (b) a "bacterium" with two flagella [15], and (c) a motor on a filament.

particles [4] where the αth particle, centered at $\mathbf{R}_\alpha$, has point forces of strength f and directions $\pm\hat{\mathbf{n}}_\alpha$ situated at $\mathbf{R}_\alpha + b\hat{\mathbf{n}}_\alpha$ and $\mathbf{R}_\alpha - b'\hat{\mathbf{n}}_\alpha$, leads to a force density

$$\mathbf{F}^a \simeq -(b+b')f\boldsymbol{\nabla}\cdot\sum_\alpha \hat{\mathbf{n}}_\alpha\hat{\mathbf{n}}_\alpha\delta(\mathbf{r}-\mathbf{R}_\alpha) + \frac{(b+b')(b-b')}{2} f\boldsymbol{\nabla}\boldsymbol{\nabla}:\sum_\alpha \hat{\mathbf{n}}_\alpha\hat{\mathbf{n}}_\alpha\hat{\mathbf{n}}_\alpha\hat{\mathbf{n}}_\alpha\delta(\mathbf{r}-\mathbf{R}_\alpha) + \ldots = \boldsymbol{\nabla}\cdot\boldsymbol{\sigma}^a, \quad (1)$$

which defines the active stress $\boldsymbol{\sigma}^a$. For bacteria swimming at speed v_0 in a fluid of viscosity η_0, $f \sim \eta_0 b v_0$. Both polar ($b \neq b'$) and apolar ($b = b'$) particles disturb the fluid; the former (a "mover") induces a nonzero fluid velocity at its center and, hence, moves, the latter (a "shaker"), by symmetry, cannot. Note that $\boldsymbol{\sigma}^a$ is insensitive, at lowest order in gradients, to the asymmetry $b - b'$: movers and shakers have the same *far-field* fluid flow, and an isotropic collection of either or both should have similar rheology. Since the force dipole determines an axis for each active particle, the natural definition $\phi(\mathbf{r})\mathbf{Q}(\mathbf{r}) \equiv \sum_\alpha(\hat{\mathbf{n}}_\alpha\hat{\mathbf{n}}_\alpha - \frac{1}{3}\mathbf{I})\delta(\mathbf{r}-\mathbf{R}_\alpha)$ (where $\mathbf{I}$ is the unit tensor) of a local nematic order parameter or alignment tensor $\mathbf{Q}$ associated with the activity lets us explore the rheological consequences of spatiotemporal correlations in the activity by a simple generalization of nematodynamics [16]. We have thus established that the active contribution to the deviatoric (traceless symmetric) stress [17]

$$\sigma^{\mathrm{a}} - (1/3)\mathbf{I}\mathrm{Tr}\,\boldsymbol{\sigma}^a = W\mathbf{Q} + W_2\mathbf{Q}^2 + \ldots, \quad (2)$$

where the constants $W, W_2 \sim (b+b')f\phi$ characterize the strength of the elementary force dipoles, and the sign of W has vital rheological consequences. The relation (2) is at the heart of the novel mechanical properties of active systems [4]. Even without equations of motion, (2) tells us why an active suspension with long-range nematic order [9] is different from its passive counterpart. Both have $\mathrm{Q} \neq 0$; the passive nematic, bound by Pascal's law since it is an equilibrium liquid despite its orientational order, has a purely *isotropic* mean stress, i.e., a pressure, whereas an active orientational ordered suspension has a nonzero mean deviatoric stress, a truly nonequilibrium effect.

Figure 2 shows what the parameter W means. In an imposed flow, in the *absence* of activity, disks (rods) tend to spend most of their time with symmetry axis along the compression (extension) axis of the flow [18]. When activity is switched on, the flow induced by the intrinsic force dipoles will clearly oppose the imposed flow in cases (a) and (b), and enhance it in (c) and (d).

For passive nematogens, $\mathbf{Q}$ is governed by a free-energy functional $F[\mathbf{Q}]$ containing polynomials in $\mathbf{Q}$ as well as Frank elastic terms $\sim\nabla\mathbf{Q}\nabla\mathbf{Q}$, giving rise to a passive order-parameter stress [18]

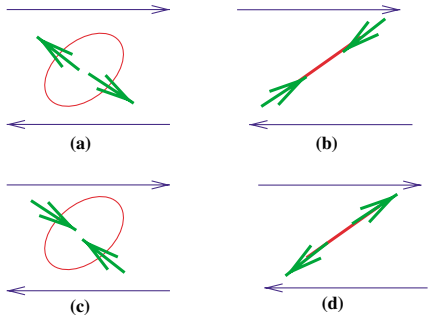

FIG. 2 (color online). Disks (a) and (c) and rods (b) and (d) with active force densities attached along their symmetry axes, under shear (horizontal arrows). The parameter $W > 0$ in (a) and (b) and <0 in (c) and (d).

$$\boldsymbol{\sigma}^{\mathrm{OP}} = 3\mathbf{G} - \mathbf{G}\cdot\mathbf{Q} - \mathbf{Q}\cdot\mathbf{G}, \quad (3)$$

where $\mathrm{G} \equiv -\delta F/\delta\mathbf{Q} + (1/3)\mathbf{I}\,\mathrm{Tr}\,\delta F/\delta\mathbf{Q}$ is the nematic molecular field. The *mean* deviatoric passive stress (3) is zero in both isotropic and nematic phases. For small nematic perturbations $\delta\mathbf{Q}$ in the isotropic phase, $F \propto a\int\phi\,\mathrm{Tr}(\delta\mathbf{Q})^2$ so that the stress fluctuation $\sim a\phi\delta\mathbf{Q}$ with a coefficient a which decreases on approaching the transition to the ordered phase. In *active* systems, the relation (2) between stress and order parameter does not arise from a free-energy functional, and the proportionality constant W has no reason to decrease with increasing nematic correlations. This difference will be seen to be crucial when we compare the pretransitional viscoelasticity of passive and active nematogenic suspensions.

Including the viscous stress $\boldsymbol{\sigma}^v = -\eta_0\mathbf{A} + O(\mathbf{Q}\boldsymbol{\nabla}\mathbf{u})$, expressed in terms of the rate of deformation $\mathrm{A} \equiv (1/2)[\boldsymbol{\nabla}\mathbf{u} + (\boldsymbol{\nabla}\mathbf{u})^T]$ and the hydrodynamic velocity field $\mathbf{u} \equiv \mathbf{g}/\rho$ for a system of density ρ, the total deviatoric stress $\boldsymbol{\sigma}$ in the active case can be written as $\boldsymbol{\sigma} = \boldsymbol{\sigma}^a + \boldsymbol{\sigma}^v + \boldsymbol{\sigma}^{\mathrm{OP}}$, plus a noise source unconstrained by a fluctuation-dissipation theorem since this is a nonequilibrium system. This defines completely the equation of motion for the momentum density $\mathbf{g}$.

The coarse-grained equation of motion for $\boldsymbol{\sigma}$ follows from that for $\mathbf{Q}$ which when *linearized* involves only terms [4] of a form present in passive nematodynamics [16]:

$$\frac{\partial\mathbf{Q}}{\partial t} = -\frac{1}{\tau}\mathbf{Q} + D\nabla^2\mathbf{Q} + \lambda_0\mathbf{A} + \cdots + \mathbf{f}, \quad (4)$$

where τ is the activity correlation time, D is a diffusivity which in passive systems would be the ratio of a Frank constant to a viscosity, λ_0 is a "reversible" kinetic coefficient [18], $\mathbf{f}$ is a traceless, symmetric, spatiotemporally white tensor noise with variance N_Q, representing thermal or active fluctuations, and the ellipsis includes the coupling of orientation to flow.

We are now ready to calculate the linear viscoelastic properties of our active suspension. In the isotropic phase, Eqs. (2)–(4), linearized and applied to spatially uniform oscillatory shear flow at frequency ω in the xy plane, imply

$$\sigma_{xy}(\omega) = -\left[\eta_0 + \frac{(a+W)\lambda_0}{-i\omega + \tau^{-1}}\right]A_{xy} \equiv -\frac{G'(\omega) - iG''(\omega)}{\omega} iA_{xy}, \quad (5)$$

which defines the storage and loss moduli $G'(\omega)$ and $G''(\omega)$. This is the claimed active enhancement or reduction $\eta_{\rm act} \propto W\tau$ of the effective viscosity at zero shear rate and zero frequency. Activity enhances viscosity in Figs. 2(a) and 2(b), since $W > 0$, and reduces it in (c) and (d) ($W < 0$). For $W > 0$, (5) tells us that the viscosity grows substantially as the system approaches a transition to orientational order (which is in general continuous for active *vectorial* order [2]), i.e., as τ is increased. By contrast, in a passive system approaching a nematic phase, the excess viscosity $\sim a\tau$ is roughly constant since $\tau \propto 1/a$.

Equation (5) also predicts strong viscoelasticity as τ increases. For *passive* systems $W = 0$. Since $a \propto \tau^{-1}$, $G'(\omega\tau \gg 1)$ decreases as $\lambda_0\eta_0/\tau$. There is little viscoelasticity near an *equilibrium* isotropic-nematic transition. For *active* systems, by contrast, W is independent of τ and of proximity to the transition. Thus, as τ grows,

$$G'(\omega\tau \gg 1) \simeq W, \quad (6)$$

independent of τ and, of course, the dynamic range over which elastic behavior is seen increases. At *equilibrium*, one would expect such strong viscoelastic behavior from a fluid or suspension near *translational* freezing, not near *orientational* ordering.

The contribution $W\mathbf{Q}$ to the deviatoric stress in active systems modifies sharply the stress vs rate flow curve. To see this, start with a passive sheared nematogenic system [19] in the isotropic phase near the transition to a nematic. Qualitatively, as the shear rate is increased from zero, $\mathbf{Q}$ increases initially linearly, then more rapidly, and then essentially linearly again, leading to shear thinning [20]. If we switch on activity, with a *positive* value of W, the rapid increase in $\mathbf{Q}$ implies an equally rapid increase in $\boldsymbol{\sigma}^a$. This will at the very least mitigate the shear thinning and, if strong enough, will lead to shear thickening. Alternatively, a system with $W < 0$ will enhance the unstable shear thinning. Note that the sign of W can be gotten from an independent experiment at low concentration, simply by seeing whether switching on activity increases or decreases the viscosity. Thus, the effect of activity on the zero frequency shear viscosity predicts the shear-thickening or shear-thinning nature of the active suspension.

We now calculate active fluctuation corrections to the shear viscosity. Equation (2) contributes an active force density $\sim W_2\nabla\mathbf{QQ}$ to the momentum equation, whose effect on viscosities, by analogy to the one-loop calculation of viscosity enhancement in model H of [21], is of the form $\Delta\eta(\omega) \sim W\int d^3k\,dt \exp(i\omega t) G_{\mathbf{Q}}(k,t) C_{\mathbf{Q}}(-k,t)$, where $G_{\mathbf{Q}}$ and $C_{\mathbf{Q}}$ are, respectively, the propagator and correlation function of $\mathbf{Q}$. From (4),

$$\frac{\Delta\eta(\omega)}{\eta_0} \sim \frac{WN_Q}{\eta_0 D^{3/2}(i\omega)^{1/2}} \qquad \text{for } \omega\tau \gg 1. \quad (7)$$

Expressing the noise strength in terms of an effective temperature $T_{\rm eff}$, and assuming on dimensional grounds $\eta_0/W \sim \tau_0$, $N_Q \sim k_B T_{\rm eff}/\tau_0$, and $D \sim \ell^2/\tau_0$, $\tau_0 = \eta_0\ell^3/k_B T_{\rm eff}$ is the rotational relaxation time of a single active particle, and ℓ its typical size, we see from (7) that $\Delta\eta(\omega)/\eta_0 \sim (\omega\tau_0)^{-1/2}$, i.e., $G^*(\omega) \sim \sqrt{i\omega}$.

All of the above effects are likely to be greatly enhanced if the transition is to a polar-ordered phase, since such a transition is expected [2] to be continuous, so that τ can increase without bound. Furthermore, since the bare time scale τ_0 is of the order of seconds for bacteria, the effects can be observed over a large dynamic range.

Activity greatly enhances the noise temperature: On dimensional grounds the variance of $\boldsymbol{\sigma}^a(k=0,\omega=0)$ is $\sim W^2\xi^3\tau$, with $W \sim \eta u_0/\xi$ for active particles moving with typical speed u_0, correlated over a scale ξ and time τ. Equating this to $k_B T_{\rm eff}\,\eta$ and estimating $\eta \sim \eta_{\rm water} = 0.01$ poise, u_0 to be a bacterial swimming speed $\sim 20\,\mu$m/s, $\tau \sim 1$ s (an *E. coli* run time) gives us a noise temperature $T_{\rm eff} \sim 10^5$–10^6 K, consistent with [22]. This will mean a thousandfold enhancement of the $t^{-d/2}$ long time tails [10] in the autocorrelation of tagged-particle velocities. On time scales shorter than τ, effects associated with spatiotemporal correlations in $\mathbf{Q}$ [23] intervene. For a drop [24] or a film [22] of size L, the tails will be cut off on a scale $\tau_v \sim \rho L^2/\pi^2\eta$. In [22], $\tau \simeq \tau_v$.

Rheology enters biology crucially through the active order parameter $\mathbf{Q}$ in several motility experiments which we discuss below. In gels, imposed strains as well as elastic anisotropies enter (4) in exactly the same way as $\mathbf{A}$ does in a fluid medium. This provides a natural explanation for *elastotaxis*, the ability of individual motile rod-shaped bacteria such as *Myxococcus xanthus* to orient with their long axes along the extension axis of an imposed elastic stress in their substrate [12], as well as *active mechanosensing* [13], where cells orient along the axis of greatest rigidity of an ambient gel. *Cytoplasmic streaming* [11], associated with the crawling of amoebae, arises naturally in our model. Figure 3 shows that an aggregate actively polymerizing at one end and depolymerizing at the other has induced flow fields with extensional and compressional axes interchanged. The resulting gradient in the active stress can be seen, in Fig. 3, to generate a mass flux from left to right. The effect will be enhanced by the fact that the depolymerizing end, with

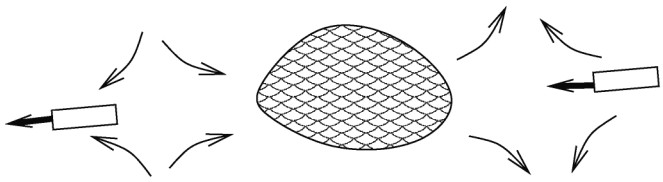

FIG. 3. Cytoplasmic streaming: Stress inhomogeneities induced by flow (thin arrows) induced in the ambient fluid due to active polymerization (at the right end) and depolymerization (at the left end) of an actin aggregate (cross-hatched region) should lead to mass flux from left to right.

negative W, is shear thinning and, hence, more fluid. These arguments suggest why such streaming always accompanies amoeboid locomotion.

In summary, we have constructed the general equations governing the rheology of suspensions of active particles, and derived several novel predictions, quantitative and qualitative. Our description is universal: Only the values of parameters such as W, τ, and λ_0 distinguish the rheologies of a bacterial suspension and a motor-microtubule extract. We look forward to tests of these predictions in experiments on living, reconstituted, or artificial [25] active-particle systems.

We thank J.-F. Joanny and J. Prost for interesting discussions and M. Fontes for bringing [12] to our attention. M. R. thanks DST, India, for a Swarnajayanti grant and the Institut Curie, Paris, for support during an enjoyable visit. S. R. acknowledges support from a Chaire Paris Science at ESPCI, Paris.

[1] See, e.g., W. Ebeling and U. Erdmann, Complexity **8**, 23 (2003); cond-mat/0307295.
[2] J. Toner and Y. Tu, Phys. Rev. E **58**, 4828 (1998).
[3] B. Alberts *et al.*, *Molecular Biology of the Cell* (Garland, New York, 2002).
[4] R. A. Simha and S. Ramaswamy, Phys. Rev. Lett. **89**, 058101 (2002).
[5] T. B. Liverpool and M. C. Marchetti, Phys. Rev. Lett. **90**, 138102 (2003).
[6] F. Nédélec, T. Surrey, and E. Karsenti, Curr. Opin. Cell Biol. **15**, 118 (2003); K. Kruse, J.-F. Joanny, F. Jülicher, J. Prost, and K. Sekimoto, Phys. Rev. Lett. **92**, 078101 (2004).
[7] F. H. C. Crick and A. F. W. Hughes, Exp. Cell Res. **1**, 37 (1950).
[8] G. N. Maksym *et al.*, J. Appl. Physiol. **89**, 1619 (2000); B. Fabry *et al.*, Phys. Rev. Lett. **87**, 148102 (2001); A. W. C. Lau *et al.*, Phys. Rev. Lett. **91**, 198101 (2003).
[9] Since active suspensions with the simplest polar or nematic order are unstable [4], these considerations hold for whatever ordered phase actually forms in such systems or else for systems stabilized by finite size.
[10] Y. Pomeau and P. Résibois, Phys. Rep. **19C**, 63 (1975).
[11] D. Bray, *Cell Movements* (Garland, New York, 2001), 2nd ed.
[12] M. Fontes and D. Kaiser, Proc. Natl. Acad. Sci. U.S.A. **96**, 8052 (1999); R. Y. Stanier, J. Bacteriol. **44**, 405 (1942).
[13] I. B. Bischofs and U. S. Schwarz, Proc. Natl. Acad. Sci. U.S.A. **100**, 9274 (2003).
[14] C. Brennen and H. Winet, Annu. Rev. Fluid Mech. **9**, 339 (1977); also see p. 385.
[15] P. R. Nott (personal communication).
[16] P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals* (Clarendon, Oxford, 1995).
[17] Active particles with more elaborate complexions of forces should give rise to the symmetry-allowed $O(\mathbf{Q}^2)$ contributions in (2).
[18] D. Forster, Phys. Rev. Lett. **32**, 1161 (1974).
[19] P. D. Olmsted and P. M. Goldbart, Phys. Rev. A **46**, 4966 (1992).
[20] P. D. Olmsted and C.-Y. D. Lu, Phys. Rev. E **56**, R55 (1997).
[21] P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. **49**, 435 (1977).
[22] X.-L. Wu and A. Libchaber, Phys. Rev. Lett. **84**, 3017 (2000).
[23] G. Grégoire, H. Chaté, and Y. Tu, Phys. Rev. E **64**, 011902 (2001).
[24] G. V. Soni *et al.*, Biophys. J. **84**, 2634 (2003)
[25] S. Nasseri and N. Phan-Thien, Comput. Mechan. **20**, 267 (1997).