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Editors' Choice

Swimmer-Microrheology

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We discuss the locomotion of a three-sphere microswimmer in a viscoelastic medium and propose a new type of active microrheology. We derive a relation that connects the average swimming velocity and the frequency-dependent viscosity of the surrounding medium. In this relation, the viscous contribution can exist only when the time-reversal symmetry is broken, whereas the elastic contribution is present only when the structural symmetry of the swimmer is broken. Purcell's scallop theorem breaks down for a three-sphere swimmer in a viscoelastic medium.

Microrheology is one of the most useful techniques for measuring the rheological properties of soft matter and various biological materials including cells.[1,2] There are two different methods: passive microrheology and active microrheology. In passive microrheology, both the local and bulk mechanical properties of a medium can be extracted from the Brownian motion of a probe particle.[3,4] In this method, the generalized Stokes–Einstein relation (GSER) is used to analyze thermal diffusive motion. In active microrheology, on the other hand, the probe is actively pulled through the fluid with the aims of driving the medium out of equilibrium and measuring mechanical responses.[5,6] Within linear response theory, the generalized Stokes relation (GSR) is employed to obtain the frequency-dependent complex shear modulus.

In this letter, we propose a new type of active microrheology using a microswimmer. Microswimmers are tiny machines that swim in a fluid such as sperm cells or motile bacteria, and are expected to be applied to microfluidics and microsystems.[7] As one of the simplest microswimmers, we consider the Najafi–Golestanian three-sphere swimmer model,[8,9] where three in-line spheres are linked by two arms of varying length (see Fig. 1). Recently, such a swimmer has been experimentally realized.[10] We investigate its motion in a general viscoelastic medium, and obtain a relation that connects the average swimming velocity and the frequency-dependent complex shear viscosity of the surrounding viscoelastic medium. We show explicitly that the absence of the time-reversal symmetry of the swimmer motion leads to the real part of the viscosity, whereas the absence of the structural symmetry of the swimmer is reflected in the imaginary part of the viscosity. Hence, we shall call the proposed method the "swimmer-microrheology". Our result also indicates that Purcell's scallop theorem,[11,12] which states that time-reversible body motion cannot be used for locomotion in a Newtonian fluid, breaks down for a three-sphere swimmer in viscoelastic media if the structural symmetry is violated.

The general equation that describes the hydrodynamics of a low-Reynolds-number flow in a viscoelastic medium is given by the following generalized Stokes equation:[13]

$$0 = \int_{-\infty}^{t} dt'\, \eta(t-t')\nabla^2 \mathbf{v}(\mathbf{r},t') - \nabla p(\mathbf{r},t). \qquad (1)$$

Here $\eta(t)$ is the time-dependent shear viscosity, $\mathbf{v}$ is the velocity field, p is the pressure field, and $\mathbf{r}$ stands for a three-dimensional positional vector. The above equation is further subjected to the incompressibility condition, $\nabla \cdot \mathbf{v} = 0$. From these equations, one can obtain a linear relation between the time-dependent force $F(t)$ acting on a hard sphere of radius a and its time-dependent velocity $V(t)$. In the Fourier domain, this relation can be represented as

$$V(\omega) = \frac{1}{6\pi\eta[\omega]a} F(\omega), \qquad (2)$$

where we use a bilateral Fourier transform for $V(\omega) = \int_{-\infty}^{\infty} dt\, V(t) e^{-i\omega t}$ and $F(\omega)$, while we employ a unilateral one for $\eta[\omega] = \int_0^{\infty} dt\, \eta(t) e^{-i\omega t}$. Equation (2) is the GSR, which has been successfully used in active microrheology experiments,[5] and its mathematical validity has also been discussed.[6]

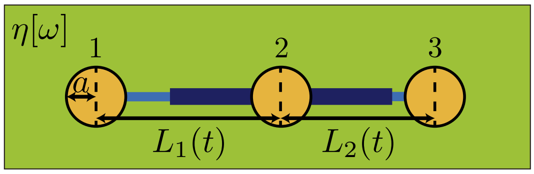

Fig. 1. (Color online) Najafi–Golestanian three-sphere swimmer model. Three identical spheres of radius a are connected by arms of lengths $L_1(t)$ and $L_2(t)$ and undergo time-dependent cyclic motion. The swimmer is embedded in a viscoelastic medium characterized by a frequency-dependent complex shear viscosity $\eta[\omega]$.

Next, we briefly explain the three-sphere model for a minimum swimmer introduced by Najafi and Golestanian.[8,9] As schematically shown in Fig. 1, this model consists of three spheres of the same radius a that are connected by two arms of lengths $L_1(t)$ and $L_2(t)$, which undergo time-dependent motion. Their explicit time dependences will be given later. If we define the velocity of each sphere along the swimmer axis as $V_i(t)$ with $i = 1, 2, 3$, we have

$$\dot{L}_1(t) = V_2(t) - V_1(t), \qquad (3)$$

$$\dot{L}_2(t) = V_3(t) - V_2(t), \qquad (4)$$

where $\dot{L}_1$ and $\dot{L}_2$ indicate time derivatives.

Owing to the hydrodynamic effect, each sphere exerts a force F_i on the viscoelastic medium and experiences a force $-F_i$ from it. To relate the forces and the velocities in the frequency domain, we use the GSR in Eq. (2) and the Oseen tensor, in which the frequency-dependent viscosity $\eta[\omega]$ is used instead of a constant one.[3,4] Assuming that $a \ll L_1, L_2$, we can write[8,9]

$$V_1(\omega) = \frac{F_1(\omega)}{6\pi\eta[\omega]a} + \frac{1}{4\pi\eta[\omega]}\frac{F_2(\omega) * L_1^{-1}(\omega)}{2\pi} + \frac{1}{4\pi\eta[\omega]}\frac{F_3(\omega) * (L_1+L_2)^{-1}(\omega)}{2\pi}, \tag{5}$$

$$V_2(\omega) = \frac{1}{4\pi\eta[\omega]}\frac{F_1(\omega) * L_1^{-1}(\omega)}{2\pi} + \frac{F_2(\omega)}{6\pi\eta[\omega]a} + \frac{1}{4\pi\eta[\omega]}\frac{F_3(\omega) * L_2^{-1}(\omega)}{2\pi}, \tag{6}$$

$$V_3(\omega) = \frac{1}{4\pi\eta[\omega]}\frac{F_1(\omega) * (L_1+L_2)^{-1}(\omega)}{2\pi} + \frac{1}{4\pi\eta[\omega]}\frac{F_2(\omega) * L_2^{-1}(\omega)}{2\pi} + \frac{F_3(\omega)}{6\pi\eta[\omega]a}, \tag{7}$$

where we have used bilateral Fourier transforms such as $L_1^{-1}(\omega) = \int_{-\infty}^{\infty} dt\,[L_1(t)]^{-1}e^{-i\omega t}$. Furthermore, the convolution of two functions is generally defined by $g_1(\omega) * g_2(\omega) = \int_{-\infty}^{\infty} d\omega'\, g_1(\omega-\omega')g_2(\omega')$ in the above equations.

As in the original study, we are interested in the autonomous net locomotion of the swimmer, and there are no external forces acting on the spheres. If the inertia of the surrounding fluid can be neglected, we have the following force balance condition:

$$F_1(t) + F_2(t) + F_3(t) = 0. \tag{8}$$

Since Eqs. (5)–(7) involve convolutions in the frequency domain, we cannot solve these equations for arbitrary $L_1(t)$ and $L_2(t)$. Here we assume that the two arms undergo the following periodic motion:

$$L_1(t) = \ell + d_1\cos(\Omega t), \tag{9}$$

$$L_2(t) = \ell + d_2\cos(\Omega t - \phi). \tag{10}$$

In the above, ℓ is the constant length, d_1 and d_2 are the amplitudes of the oscillatory motion, Ω is the common arm frequency, and ϕ is the mismatch in the phases between the two arms. In the following analysis, we generally assume that $d_1, d_2 \ll \ell$. The *time-reversal symmetry* of the arm motion is present when $\phi = 0$ and π. Furthermore, we characterize the *structural symmetry* of the swimmer by d_1 and d_2, i.e., the structure is symmetric when $d_1 = d_2$, while it is asymmetric when $d_1 \neq d_2$.

Since the arm frequency is Ω, we assume that the velocities and the forces of the three spheres can generally be written as

$$V_i(\omega) = V_{i,0}\,\delta(\omega) + \sum_{n=1}^{\infty}[V_{i,n}\,\delta(\omega+n\Omega) + V_{i,-n}\,\delta(\omega-n\Omega)], \tag{11}$$

$$F_i(\omega) = F_{i,0}\,\delta(\omega) + \sum_{n=1}^{\infty}[F_{i,n}\,\delta(\omega+n\Omega) + F_{i,-n}\,\delta(\omega-n\Omega)], \tag{12}$$

where $i = 1, 2, 3$ for the three spheres. Substituting Eqs. (11) and (12) into the six coupled Eqs. (3)–(8), we obtain a matrix equation with infinite dimensions.

Under the conditions $d_1, d_2 \ll \ell$ and $a \ll \ell$, we are allowed to consider only $n = -1, 0, 1$ and we further use the approximation $F_{i,\pm 2} \approx 0$. Then we can solve for the six unknown functions $V_i(\omega)$ and $F_i(\omega)$, and also calculate the total swimming velocity $V = (V_1 + V_2 + V_3)/3$. Up to the lowest order terms in a, the average swimming velocity over one cycle of motion becomes[14)]

$$\overline{V} \approx \frac{7d_1d_2a\Omega}{24\ell^2}\frac{\eta'[\Omega]}{\eta_0}\sin\phi - \frac{5(d_1^2 - d_2^2)a\Omega}{48\ell^2}\frac{\eta''[\Omega]}{\eta_0}, \tag{13}$$

where $\eta'[\Omega]$ and $\eta''[\Omega]$ are the real and imaginary parts of the complex shear viscosity, respectively, and $\eta_0 = \eta[\Omega \to 0]$ is the constant zero-frequency viscosity.

Table I. Locomotion of a three-sphere swimmer in a viscoelastic medium and the relevant rheological information.

Medium	Viscous				Viscoelastic			
Time-reversal symmetry	Y		N		Y		N	
Structural symmetry	Y	N	Y	N	Y	N	Y	N
Swimmer motion	N	N	Y	Y	N	Y	Y	Y
Rheological information	—	—	N	N	—	η''	η'	η', η''

The first term in Eq. (13) can be regarded as the viscous contribution and is present only if the time-reversal symmetry of the swimmer motion is broken, i.e., $\phi \neq 0, \pi$. The second term, on the other hand, corresponds to the elastic contribution, and exists only when the structural symmetry of the swimmer is broken, i.e., $d_1 \neq d_2$. If we are able to control d_1, d_2, and Ω of the swimmer, we can first obtain $\eta'[\Omega]$ by measuring $\overline{V}$ as a function of Ω by setting $d_1 = d_2$. Then we make a difference between d_1 and d_2 to examine the change in $\overline{V}$, which then yields $\eta''[\Omega]$. The corresponding complex shear modulus is simply obtained by $G[\Omega] = i\Omega\eta[\Omega]$. This is a new type of active microrheology that we propose in this letter.

For a purely Newtonian fluid, namely, for a medium characterized by a constant viscosity, the second term in Eq. (13) vanishes, and the first term coincides with the expression obtained by Golestanian and Ajdari.[9)] It should be noted here, however, that the velocity $\overline{V}$ in this case no longer depends on the constant viscosity (because $\eta'[\Omega]/\eta_0 = 1$) and we cannot measure it from $\overline{V}$. Equation (13) also implies that the swimmer cannot move in a purely elastic medium, for which we have $\eta_0 \to \infty$. Importantly, owing to the presence of the second term, Purcell's scallop theorem breaks down for a three-sphere swimmer in a viscoelastic medium. Namely, even if the time-reversal symmetry of the swimmer motion is not broken, i.e., $\phi = 0, \pi$, the present swimmer can still move in a viscoelastic medium due to the second term as long as its structural symmetry is broken, i.e., $d_1 \neq d_2$. On the basis of Eq. (13), we have summarized in Table I the motion of a three-sphere swimmer in a viscoelastic medium and the relevant rheological information.

To further illustrate our result, we first assume that the surrounding viscoelastic medium is described by a simple Maxwell model. In this case, the frequency-dependent viscosity can be written as

$$\eta[\omega] = \eta_0\frac{1 - i\omega\tau_M}{1 + \omega^2\tau_M^2}, \tag{14}$$

where τ_M is the characteristic time scale. Within this model, the medium behaves as a viscous fluid for $\omega\tau_M \ll 1$, while it becomes elastic for $\omega\tau_M \gg 1$. Using Eq. (14), we can easily obtain the average swimming velocity in Eq. (13) as

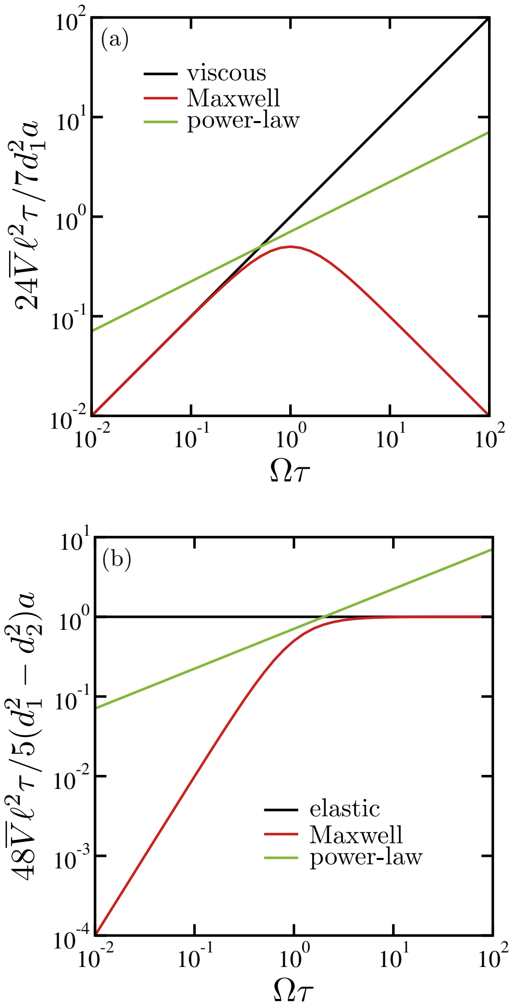

Fig. 2. (Color online) Average swimming velocity $\overline{V}$ as a function of $\Omega\tau$, where Ω is the arm frequency and τ represents either $\tau_{\rm M}$ for a Maxwell fluid (red lines) or $\tau_{\rm p}$ for a power-law fluid (green lines). In the power-law model, we choose $\alpha = 1/2$. (a) Viscous contribution by setting $\phi = \pi/2$ and $d_1 = d_2$. Here $\overline{V}$ is scaled by $7d_1^2 a/(24\ell^2\tau)$. The case for a viscous fluid is plotted by the black line. (b) Elastic contribution by setting $\phi = 0$ and $d_1 \neq d_2$. Here $\overline{V}$ is scaled by $5(d_1^2 - d_2^2)a/(48\ell^2\tau)$. The case for an elastic medium is plotted by the black line.

$$\overline{V} = \frac{7d_1 d_2 a\Omega}{24\ell^2}\frac{1}{1+\Omega^2\tau_{\rm M}^2}\sin\phi + \frac{5(d_1^2 - d_2^2)a\Omega}{48\ell^2}\frac{\Omega\tau_{\rm M}}{1+\Omega^2\tau_{\rm M}^2}. \quad (15)$$

The first viscous term increases as $\overline{V} \sim \Omega$ for $\Omega\tau_{\rm M} \ll 1$, while it decreases as $\overline{V} \sim \Omega^{-1}$ for $\Omega\tau_{\rm M} \gg 1$. This is a unique feature of the viscoelasticity,[7,15,16] but such a reduction occurs simply because the medium responds elastically in the high-frequency regime. On the other hand, the second elastic term increases as $\overline{V} \sim \Omega^2$ for $\Omega\tau_{\rm M} \ll 1$, and it approaches a constant for $\Omega\tau_{\rm M} \gg 1$. In Fig. 2(a), we plot the average swimming velocity $\overline{V}$ as a function of the dimensionless arm frequency $\Omega\tau_{\rm M}$ when $\phi = \pi/2$ and $d_1 = d_2$. This plot corresponds to the first term in Eq. (15). As a reference, the behavior of $\overline{V} \sim \Omega$ for a purely viscous fluid is also plotted. Figure 2(b) is a similar plot when $\phi = 0$ and $d_1 \neq d_2$, and corresponds to the second term in Eq. (15).

As a different example, we next consider the case in which the viscoelastic medium is described by a power-law model such that[13,17,18)]

$$\eta[\omega] = G_0(i\omega)^{\alpha-1}, \quad (16)$$

where the exponent can take values of $0 \le \alpha \le 1$. With this expression, the complex shear modulus also exhibits a power-law behavior, $G[\omega] = G_0(i\omega)^{\alpha}$. The limits of $\alpha = 0$ and 1 correspond to the purely elastic and the purely viscous cases, respectively. In the case of a power-law fluid, the average swimming velocity can be obtained from Eqs. (13) and (16) as

$$\overline{V} = \frac{7d_1 d_2 a}{24\ell^2\tau_{\rm p}}(\Omega\tau_{\rm p})^{\alpha}\sin(\pi\alpha/2)\sin\phi + \frac{5(d_1^2 - d_2^2)a}{48\ell^2\tau_{\rm p}}(\Omega\tau_{\rm p})^{\alpha}\cos(\pi\alpha/2), \quad (17)$$

where $\tau_{\rm p} = (\eta_0/G_0)^{1/(1-\alpha)}$. Here we have assumed that the medium behaves as a purely viscous fluid in the low-frequency limit characterized by a finite viscosity η_0. According to the above expression, the swimming velocity scales as $\overline{V} \sim \Omega^{\alpha}$ in both the first and second terms. For the purely viscous case of $\alpha = 1$, the first term reduces to the result by Golestanian and Ajdari,[9] while the second term vanishes. For the purely elastic case of $\alpha = 0$, on the other hand, the first term vanishes and the second term remains, although the latter no longer depends on the arm frequency Ω. In Figs. 2(a) and 2(b), we have also plotted the average velocity $\overline{V}$ as a function of $\Omega\tau_{\rm p}$ when $\alpha = 1/2$. In both of these plots, the scaling behavior $\overline{V} \sim \Omega^{1/2}$ is seen.

Lauga considered the axisymmetric squirming motion of a sphere (squirmer) embedded in an Oldroyd-B fluid, which represents a typical polymeric fluid.[19] He reported that the scallop theorem in a viscoelastic fluid breaks down if the squirmer has fore-aft asymmetry in its surface velocity distribution. For a time-reversal deformation given by a simple sinusoidal gait, he showed that the average swimming velocity is given by $\overline{V} \sim \Omega\,{\rm De}/(1+{\rm De}^2)$, where the Deborah number is given by ${\rm De} = \Omega\tau_{\rm O}$ with a characteristic relaxation time $\tau_{\rm O}$ in the Oldroyd-B model. Such a frequency dependence of the swimming velocity is identical to the second term of Eq. (15) obtained for a Maxwell fluid, although Eq. (13) is more general. On the other hand, our result is different from that by Curtis and Gaffney,[20] because they showed that the swimming velocity in a viscoelastic medium is the same as that in a Newtonian fluid.

To summarize, we have proposed a new active microrheology using the Najafi–Golestanian three-sphere swimmer. The frequency dependence of the average swimming speed provides us with the complex shear viscosity of the surrounding viscoelastic medium. Here the viscous contribution can exist only when the time-reversal symmetry of the swimmer is broken, whereas the elastic contribution is present only if its structural symmetry is broken.

Even though the argument in this Letter is restricted to the artificial three-sphere swimmer, we expect that our basic concept can still be applied to more complex biological processes such as the motion of bacteria, flagellated cellular swimming, and the beating of cilia. Since most of these phenomena take place in a viscoelastic environment, we hope that the concept of our new active microrheology will be used in the future to reveal their mechanical and dynamical properties.

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1) T. M. Squires and T. G. Mason, Annu. Rev. Fluid Mech. **42**, 413 (2010).
2) D. T. N. Chen, Q. Wen, P. A. Janmey, J. C. Crocker, and A. G. Yodh, Annu. Rev. Condens. Matter Phys. **1**, 301 (2010).
3) T. G. Mason and D. A. Weitz, Phys. Rev. Lett. **74**, 1250 (1995).
4) T. G. Mason, Rheol. Acta **39**, 371 (2000).
5) F. Gittes, B. Schnurr, P. D. Olmsted, F. C. MacKintosh, and C. F. Schmidt, Phys. Rev. Lett. **79**, 3286 (1997).
6) B. Schnurr, F. Gittes, F. C. MacKintosh, and C. F. Schmidt, Macromolecules **30**, 7781 (1997).
7) E. Lauga and T. R. Powers, Rep. Prog. Phys. **72**, 096601 (2009).
8) A. Najafi and R. Golestanian, Phys. Rev. E **69**, 062901 (2004).
9) R. Golestanian and A. Ajdari, Phys. Rev. E **77**, 036308 (2008).
10) G. Grosjean, M. Hubert, G. Lagubeau, and N. Vandewalle, Phys. Rev. E **94**, 021101(R) (2016).
11) E. M. Purcell, Am. J. Phys. **45**, 3 (1977).
12) E. Lauga, Soft Matter **7**, 3060 (2011).
13) R. Granek, Soft Matter **7**, 5281 (2011).
14) (Supplemental Materials) The detailed derivation of Eq. (13) is provided online.
15) H. C. Fu, T. R. Powers, and C. W. Wolgemuth, Phys. Rev. Lett. **99**, 258101 (2007).
16) H. C. Fu, C. W. Wolgemuth, and T. R. Powers, Phys. Fluids **21**, 033102 (2009).
17) S. Komura, S. Ramachandran, and K. Seki, EPL **97**, 68007 (2012).
18) S. Komura, K. Yasuda, and R. Okamoto, J. Phys.: Condens. Matter **27**, 432001 (2015).
19) E. Lauga, EPL **86**, 64001 (2009).
20) M. P. Curtis and E. A. Gaffney, Phys. Rev. E **87**, 043006 (2013).

