

Odd Mobility of a Passive Tracer in a Chiral Active Fluid

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(Received 19 November 2022; accepted 16 March 2023; published 12 April 2023)

Chiral active fluids break both time-reversal and parity symmetry, leading to exotic transport phenomena unobservable in ordinary passive fluids. We develop a generalized Green-Kubo relation for the anomalous lift experienced by a passive tracer suspended in a two-dimensional chiral active fluid subjected to an applied force. This anomalous lift is characterized by a transport coefficient termed the odd mobility. We validate our generalized response theory using molecular dynamics simulations, and we show that the asymmetric tracer mobility may be understood mechanically in terms of asymmetric deformations of the tracer-fluid density distribution function. We show that the even and odd components of the mobility decay at different rates with tracer size, suggesting the possibility of size-based particle separation using a chiral active working fluid.

DOI: [10.1103/PhysRevLett.130.158201](https://doi.org/10.1103/PhysRevLett.130.158201)

Chiral active fluids are composed of constituents that convert energy into directed rotational motion [1–3]. The quiescent state of such a fluid breaks both parity and time-reversal symmetry and accordingly is not a state of thermodynamic equilibrium. These broken symmetries lead to a variety of exotic transport phenomena [1–15]. For example, an odd viscosity that links dilational flow to shear stress and sheared flow to normal stress can translate forcing into fluid motion in an orthogonal direction [4,9,10]. Describing transport phenomena in such non-equilibrium fluids is challenging because classical linear response theories are valid perturbatively only about an equilibrium steady state [16]. Here, we consider a similar transport process, the motion of a passive tracer suspended in a two-dimensional, odd viscous chiral active fluid and subjected to a constant force, and we develop a response theory based on ensembles of trajectories [17,18]. Recent work has examined the advective [13] and driven [8] transport of passive tracers in odd viscous fluids. These theoretical treatments are phenomenological, however, illustrating the need for a microscopic description of tracer mobility in chiral active fluids. The framework employed here can be extended to arbitrary order in the applied forcing and is valid for a reference state arbitrarily far from equilibrium [19].

In general, the velocity response of a passive tracer is characterized by a second rank tensor called the mobility tensor by the relation $\langle \mathbf{V} \rangle_F = \boldsymbol{\mu} \cdot \mathbf{F}$, where $\mathbf{V}$ is the tracer velocity, $\boldsymbol{\mu}$ is the mobility tensor, $\mathbf{F}$ is an external force applied to the tracer, and angled brackets denote a non-equilibrium ensemble average [7,8]. In passive fluids, isotropy requires that the mobility tensor be proportional

to the unit tensor, introducing a single transport coefficient μ_e , termed the even mobility. However, in two-dimensional fluids breaking parity and time-reversal symmetry, the isotropy of the second rank two-dimensional Levi-Civita tensor permits the emergence of an additional transport coefficient μ_o , termed the odd mobility, analogous to the emergence of the odd viscosity [4]. The general expression for the mobility tensor is given in Cartesian coordinates by

$$\boldsymbol{\mu} = \mu_e \mathbf{1} + \mu_o \boldsymbol{\epsilon} = \begin{pmatrix} \mu_e & \mu_o \\ -\mu_o & \mu_e \end{pmatrix}, \quad (1)$$

where $\mathbf{1}$ is the unit tensor, and $\boldsymbol{\epsilon}$ is the Levi-Civita tensor. Under a force $\mathbf{F} = F\hat{x}$ applied in the positive x direction, the resulting tracer velocity is $\langle \mathbf{V} \rangle_F = \mu_e F\hat{x} - \mu_o F\hat{y}$, indicating that the particle experiences an anomalous lift proportional to the odd mobility in the direction orthogonal to the applied force. This lift corresponds to the conversion of directed rotational motion of the underlying fluid into a uniform deflection of the tracer particle. Such a behavior could be observed experimentally by subjecting tracers in an odd viscous fluid [15] to an external force.

Here, we consider odd mobility numerically with a simple model containing a passive Weeks-Chandler-Andersen (WCA) particle [20] suspended in a fluid of active dimers [2]. The dimers are composed of monomers linked by stiff harmonic bonds that interact with each other and the passive tracer via a purely repulsive WCA potential characterized by an energy parameter $\beta\epsilon = 1$, where $\beta \equiv 1/k_B T$ is the inverse of the temperature times Boltzmann's constant. Each monomer is also subjected to an active force of constant magnitude F_a , directed perpendicular to the

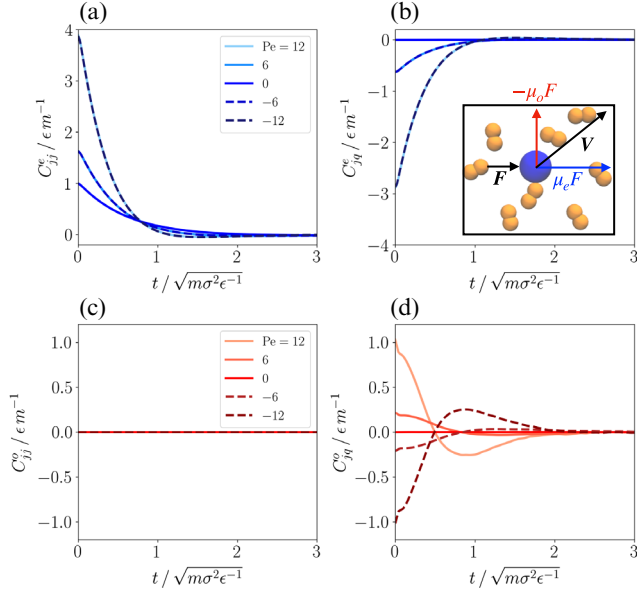


FIG. 1. Correlation functions contributing to the even and odd mobility response of a passive tracer for several different Péclet numbers and density $\bar{n} = 0.2\sigma^{-2}$. (a) The current autocorrelation for the even mobility. (b) The current-frenesy cross-correlation for the even mobility. (c) and (d) The current autocorrelation (c) and current-frenesy cross-correlation for the odd mobility. The inset in (b) shows a simulation snapshot and the decomposition of the velocity response of a passive tracer subjected to an applied force in terms of the even and odd mobility coefficients. The blue particle is the passive tracer, and the orange particles are the actively rotating dimers. Errors are smaller than the line thickness.

instantaneous dimer bond. This active force results in a directed active torque experienced by each dimer of average magnitude $F_a d_0$, where $d_0 = \sigma$ is the equilibrium bond length set equal to the monomer diameter σ . The active forces are oriented such that $F_a > 0$ corresponds to a counterclockwise active torque. The monomers undergo underdamped Langevin dynamics with the equation of motion

$$m\dot{\mathbf{v}}_i^\alpha = -\gamma\mathbf{v}_i^\alpha + \mathbf{F}_{c,i}^\alpha + \mathbf{F}_{a,i}^\alpha + \sqrt{2\gamma/\beta}N_i^\alpha(t), \quad (2)$$

where i is the dimer index and $\alpha \in \{1, 2\}$ indexes the monomers on dimer i , m is monomer mass, $\mathbf{F}_{c,i}^\alpha$ are the conservative forces acting on monomer $i - \alpha$, $\mathbf{F}_{a,i}^\alpha$ is the active force, and γ is a linear friction coefficient describing dissipation from the particles to an underlying quiescent fluid bath. (See Supplemental Material [21], which includes Refs. [22,23].) $N_i(t)$ is a delta-correlated Gaussian fluctuating force with mean zero and unit variance. A simulation snapshot is shown in the inset in Fig. 1(a), and details of the dimer model can be found in Refs. [2,24]. This model represents a minimal model of a chiral active fluid

exhibiting odd viscosity [2]. The Supplemental Material contains further simulation details [21].

The tracer equation of motion is

$$m\dot{\mathbf{V}} = -\gamma\mathbf{V} + \mathbf{F}_{\text{WCA}} + \mathbf{F} + \sqrt{2\gamma/\beta}\mathbf{N}(t), \quad (3)$$

where $\mathbf{V} \equiv \dot{\mathbf{X}}$ is the tracer velocity, $\mathbf{X}$ is the tracer position, $\mathbf{F}_{\text{WCA}}$ is the total WCA force on the tracer from the surrounding monomers, and $\mathbf{F}$ is a constant external force. We take the tracer mass and friction coefficient equal to that for the monomers in all simulations. Except in Fig. 2(c), we also take the tracer diameter σ_{tracer} to be equal to σ . Though it does not appear explicitly in (3), the dimer activity has a profound influence on the tracer dynamics. This effect is contained in the average structure of $\mathbf{F}_{\text{WCA}}$, as we will explore below.

The Gaussian nature of the noise allows us to write the path probability density as [25]

$$\mathbb{P}_{\mathbf{F}}[X] \propto e^{-\beta S_{\mathbf{F}}[X]}, \quad (4)$$

where $X \equiv \{\mathbf{X}(t)\}_{t=0}^{t_N}$ is a tracer trajectory of duration $t_N \rightarrow \infty$, and the path action under an applied force $\mathbf{F}$ is given by the Onsager-Machlup construction [26,27] as

$$\begin{aligned} S_{\mathbf{F}}[X] &= \frac{1}{4\gamma} \int_0^{t_N} dt (m\dot{\mathbf{V}} + \gamma\mathbf{V} - \mathbf{F}_{\text{WCA}} - \mathbf{F})^2 \\ &\equiv S_0[X] + \Delta S_{\mathbf{F}}[X]. \end{aligned} \quad (5)$$

In the second equality, we have extracted the unbiased path action $S_0[X]$ obtained when $\mathbf{F} = 0$. The relative path action $\Delta S_{\mathbf{F}}[X]$ is defined by

$$\frac{\mathbb{P}_{\mathbf{F}}[X]}{\mathbb{P}_0[X]} = e^{+\beta \Delta S_{\mathbf{F}}[X]}, \quad (6)$$

with

$$\Delta S_{\mathbf{F}}[X] \approx \frac{1}{2} \mathbf{J} \cdot \mathbf{F} + \frac{1}{2} \mathbf{Q} \cdot \mathbf{F}, \quad (7)$$

where we have defined the time-extensive current $\mathbf{J} \equiv \int dt \mathbf{V}$ and frenesy $\mathbf{Q} \equiv -\int dt (\mathbf{F}_{\text{WCA}}/\gamma)$ [28]. We have retained terms only to first order in $\mathbf{F}$ as we are interested only in the linear response, and we have dropped the temporal boundary term proportional to $\int dt m\dot{\mathbf{V}}$ because its contribution to the mobility exactly vanishes in the limit $t_N \rightarrow \infty$. (See Supplemental Material [21].)

Equation (6) allows us to reweight path ensemble averages taken in a forced ($\mathbf{F} \neq 0$) ensemble to those in an unforced one, which we may use to develop a linear response expression for the mobility. Rewriting the trajectory average of $\mathbf{J}$ in the presence of $\mathbf{F}$ as one in its absence using Eq. (6) and expanding to first order in $\mathbf{F}$, we obtain

$$\langle \delta \mathbf{J} \rangle_{\mathbf{F}} = t_N \langle \mathbf{V} \rangle_{\mathbf{F}} = \frac{\beta}{2} (\langle \delta \mathbf{J} \otimes \delta \mathbf{J} \rangle_0 + \langle \delta \mathbf{J} \otimes \delta \mathbf{Q} \rangle_0) \cdot \mathbf{F}, \quad (8)$$

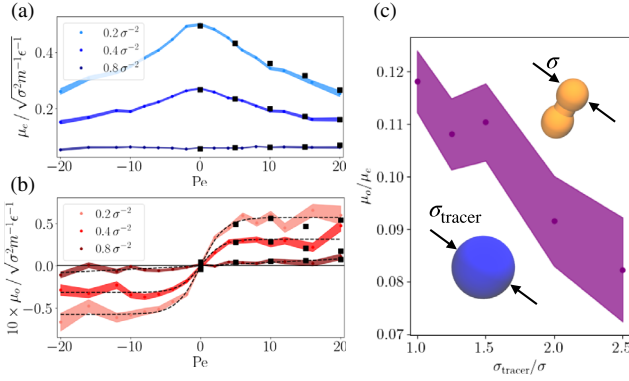


FIG. 2. Mobility as a function of activity, density, and tracer diameter. (a),(b) Even and odd mobility as a function of Péclet number for three different densities. (c) Ratio of odd to even mobility as a function of tracer diameter for $\bar{n} = 0.2\sigma^{-2}$. Shaded regions indicate standard errors. The black squares in (a) and (b) show the results of forced mobility calculations. The dashed black lines in (b) are guides to the eye.

where $\delta\mathcal{O} \equiv \mathcal{O} - \langle\mathcal{O}\rangle_0$. We insert our definition for the mobility tensor $\langle\mathbf{V}\rangle = \boldsymbol{\mu} \cdot \mathbf{F}$ and differentiate with respect to $\mathbf{F}$ to obtain a linear response relationship for the mobility tensor directly,

$$\boldsymbol{\mu} = \frac{\beta}{2t_N} (\langle\delta\mathbf{J} \otimes \delta\mathbf{J}\rangle_0 + \langle\delta\mathbf{J} \otimes \delta\mathbf{Q}\rangle_0). \quad (9)$$

This equation represents the key theoretical result of this Letter. The first term on the right-hand side of Eq. (9) contains only the steady-state fluctuations in the current and corresponds to the ordinary “thermodynamic” response obtained about equilibrium steady states. The second term contains the current-frenesy correlation and is nonvanishing only in nonequilibrium reference states. Physically, the frenesy represents microscopic motion unassociated with the current. Though the particular form of the frenesy depends on the nature of the underlying microscopic dynamics [29–31], for tracer motion well described by the underdamped Langevin equation given in Eq. (3), the frenesy requires knowledge only of the forces and their time correlations. Therefore, in principle, it may be measured experimentally for such a system.

We can make explicit the time integrals appearing in the definitions of $\mathbf{J}$ and $\mathbf{Q}$ in Eq. (9). Using time-translation invariance and averaging the diagonal and off-diagonal components of the mobility tensor, we find

$$\mu_e = \beta \int_0^\infty dt [C_{jj}^e(t) + C_{jq}^e(t)], \quad (10)$$

and

$$\mu_o = \beta \int_0^\infty dt C_{jq}^o(t), \quad (11)$$

with

$$C_{jj}^e(t) \equiv \frac{1}{2} \sum_{\alpha=x,y} \langle\delta j_\alpha(t) \delta j_\alpha(0)\rangle_0, \quad (12)$$

$$C_{jq}^e(t) \equiv \frac{1}{4} \sum_{\alpha=x,y} [\langle\delta j_\alpha(t) \delta q_\alpha(0)\rangle_0 + \langle\delta q_\alpha(t) \delta j_\alpha(0)\rangle_0], \quad (13)$$

and

$$C_{jq}^o(t) \equiv \frac{1}{4} [\langle\delta j_x(t) \delta q_y(0)\rangle_0 + \langle\delta q_y(t) \delta j_x(0)\rangle_0] - \frac{1}{4} [\langle\delta j_y(t) \delta q_x(0)\rangle_0 + \langle\delta q_x(t) \delta j_y(0)\rangle_0]. \quad (14)$$

In the above, $\mathbf{j} \equiv \mathbf{V}$, and $\mathbf{q} \equiv -\mathbf{F}_{\text{WCA}}/\gamma$. The contribution of the current autocorrelation to the odd mobility exactly vanishes. This can be seen clearly from Eq. (9) where the thermodynamic contribution to the mobility is proportional to the explicitly symmetric variance of the time-extensive current.

Figure 1 shows the contributions of each correlation function to the mobilities. We plot the results for several different activities, quantified by a Péclet number $\text{Pe} \equiv 2\beta F_a d_0$. We include the results for equilibrium, $\text{Pe} = 0$, for which the odd mobility must vanish. Indeed, we observe that C_{jq}^o is zero. For finite Pe , both the current (thermodynamic) and current-frenesy (frenetic) contributions to μ_e are invariant under change of sign of Pe . This is a reflection of the fact that μ_e enters into the dissipation, $\langle\mathbf{V}\rangle_{\mathbf{F}} \cdot \mathbf{F} = \mu_e F^2$, and hence is constrained to remain positive under time reversal. It therefore must be invariant under parity inversion, $F_a \rightarrow -F_a$, in order to preserve overall parity-time symmetry.

The initial values of the thermodynamic and frenetic correlation functions for the even mobility both grow with increasing magnitude Pe , though the current variance grows more rapidly. Adding these curves shows that the growth of the frenetic contribution acts to maintain a constant initial value of the overall correlation function, while the relatively larger frenetic integral relaxation time results in a decreasing overall relaxation time with increasing activity and hence a decreasing even mobility. (See Supplemental Material [21].) This is consistent with the decrease in overall even mobility with increasing activity shown in Fig. 2(a).

The thermodynamic contribution to the odd mobility C_{jj}^o exactly vanishes for all activities. As noted above, this is predicted analytically and reflects the fact that the odd mobility is a purely nonequilibrium phenomenon. The frenetic correlation functions show a rich structure, possessing an initial shift in their decay rate at short times and a change in sign at longer times. The curves are inverted under a change in sign of F_a , reflecting the parity—and hence time inversion—antisymmetry of the odd mobility.

This is permitted because the odd mobility does not contribute directly to the tracer dissipation, analogous to the odd viscosity of the underlying fluid [4].

In Fig. 2, we show the even and odd mobility responses as measured by Eq. (9) as a function of activity for several different densities. As anticipated, both the even and odd mobility responses are suppressed as density is increased. The maximum ratio of odd to even mobilities is obtained for the lowest density examined, $\bar{n} = 0.2\sigma^{-2}$, and is roughly 20% for $|\text{Pe}| = 20$. Surprisingly, we find that the even mobility is maximized for $\text{Pe} = 0$ and decreases with increasing activity, demonstrating that a simple understanding of the activity in terms of an effective temperature does not hold in this system, as previous results have reported an increase in the mean squared diffusivity with Pe [7].

At the highest density, $\bar{n} = 0.8\sigma^{-2}$, the odd mobility is observed to vary linearly over the range of Péclet numbers examined, with positive values, corresponding to downward deflections under a force applied in the positive x direction, obtained for counterclockwise dimer rotations and vice versa. However, we find that at lower densities the odd mobility appears to saturate to a constant value as $|\text{Pe}|$ is increased. Combined with the observed monotonically decreasing nature of μ_e with increasing $|\text{Pe}|$, this indicates that the ratio of odd to even mobility may be made arbitrarily large for high activity, suggesting the possibility of a regime in which the particle is deflected by essentially 90° from the applied force. We note that in Figs. 2(a) and 2(b) we have compared our simulation results in the reference steady state to runs in which a force is applied directly to the tracer particle and its velocity is directly measured, and the results are in excellent agreement.

Finally, in Fig. 2(c), we plot the ratio of even to odd mobility as a function of tracer diameter σ_{tracer} . We plot this ratio only for a single Pe as we observe that this curve is essentially independent of Pe for finite activity. We find that the ratio of mobilities decreases with increasing σ_{tracer} , indicating that μ_o decreases more rapidly than μ_e as tracer size is increased. The angle θ between applied force and resultant tracer velocity is given by $\tan \theta = -\mu_o/\mu_e$ [32,33], indicating that the particle deflection angle is therefore size dependent. This suggests the possibility of size-based particle separation using an odd viscous fluid as a working fluid.

We can understand the origin of the trends in mobility mechanically by deriving a relationship between the tracer-monomer pair distribution function (PDF) and the mobility response [30,31,34]. Averaging the tracer equation of motion (3) over the noise, we obtain

$$\langle \mathbf{V} \rangle_F = \frac{1}{\gamma} (\mathbf{F} + \langle \mathbf{F}_{\text{WCA}} \rangle_F). \quad (15)$$

Given the pairwise nature of the repulsive interaction, the average of the components of the velocity

parallel and orthogonal to the applied force can be written as

$$V_{\parallel} = \frac{F}{\gamma} + \frac{\bar{n}}{\gamma} \int d\mathbf{x} g(\mathbf{x}|\mathbf{F}) F_{\text{WCA},\parallel}(\mathbf{x}), \quad (16)$$

and

$$V_{\perp} = \frac{\bar{n}}{\gamma} \int d\mathbf{x} g(\mathbf{x}|\mathbf{F}) F_{\text{WCA},\perp}(\mathbf{x}), \quad (17)$$

respectively, where $g(\mathbf{x}|\mathbf{F})$ is the tracer-monomer PDF in the presence of an applied force $\mathbf{F}$ [35,36], and $F_{\text{WCA}}(\mathbf{x})$ is the force field characterizing the WCA interaction between monomers and the tracer particle held fixed at $\mathbf{X} = 0$.

We take $\mathbf{F}$ to be directed along the positive x direction. Intuitively, we anticipate that a relative decrease in $V_{\parallel}$ —and hence μ_e —with increasing $|\text{Pe}|$ will be associated with an increase in monomer density accumulated on the front of the particle. However, we observe the opposite in Figs. 3(a) and 3(b), where we plot the PDF for $\text{Pe} = 0$ and 20 under an applied force. Instead, we observe that the ridge of monomer density accumulated on the front of the tracer decreases substantially in magnitude with increasing activity, ostensibly suggesting that μ_e should increase with increasing magnitude Pe —the opposite of what is observed in Fig. 2. This apparent inconsistency is resolved by considering the relative density distribution.

Though the accumulation of monomer density in front of the tracer decreases with increasing activity, a high density ridge is pushed further into the interaction region with the

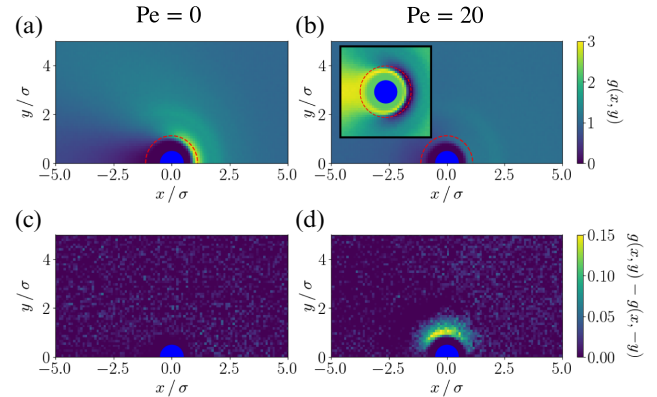


FIG. 3. Tracer-monomer PDF for a density of $\bar{n} = 0.2\sigma^{-2}$ and different Péclet numbers under a force applied to the tracer in the positive x direction of magnitude $F = 10\epsilon\sigma^{-1}$. The left and right columns correspond to Péclet numbers of (a),(c) 0 and (b),(d) 20. The upper row shows the PDF in the upper half plane, while (c), (d) shows the PDF in the upper half plane minus the PDF in the lower half plane, illustrating the reflection asymmetry in y . The solid blue half circle indicates the location of the tracer particle. The inset in (b) shows the difference in PDFs obtained at $\text{Pe} = 20$ and 0, and the dashed red line in (a), (b), and the inset indicates the cutoff of the WCA interaction.

tracer, resulting in a much stronger resistive interaction. This is clearly illustrated in the inset in Fig. 3(b), which shows the PDF obtained for $Pe = 20$ less than that obtained for $Pe = 0$. Direct integration confirms that this shift in particle distribution results in the suppression of the even mobility at finite activity. Interestingly, this shift in the high density ridge is not observed in the PDF calculated using the dimer center of mass coordinates, indicating that the dimers accumulated at the front of the tracer are preferentially oriented with a component of their bond vector orthogonal to the face of the tracer, and that this “orientational locking” increases with activity. This mechanism is fundamentally linked to the biased nature of the rotational motion of the dimers and is enhanced with greater activity. It therefore cannot be renormalized through an effective temperature that implicitly treats the effect of the activity as enhanced unbiased random noise of the dimer particles.

Given the central nature of the WCA interaction, $F_{WCA,\perp}$ is necessarily antisymmetric in y . Therefore, the deflection associated with the odd mobility must be the result of an asymmetry in the PDF under an applied force for finite Pe such that the integral in Eq. (17) is nonvanishing. Indeed, this is observed in Figs. 3(c) and 3(d), where we plot the PDF in the upper half plane less than in the lower half plane, $g(x, y) - g(x, -y)$. This is a direct measure of the asymmetry under the transformation $y \rightarrow -y$ and hence of a nonvanishing odd mobility. We observe that there is measurable asymmetry only in the case of finite Péclet number, and the odd mobility is therefore due to asymmetric deformations of the particle distribution about the tracer particle for finite activities.

In this Letter, we have derived a generalized Green-Kubo relation relating current and frenesy fluctuations in a steady state far from equilibrium to the mobility of a passive tracer suspended in a two-dimensional chiral active fluid. We have validated our results using molecular dynamics simulations and have shown that, whereas the even mobility counterintuitively decreases with increasing activity, the odd mobility increases until it saturates to a finite value. The former result indicates that an effective temperature description of the mobility response is invalid in this system. We have further shown that the odd mobility decays more rapidly than the even mobility with increasing tracer particle size, independent of activity, indicating the possibility of size-based particle separation using an odd viscous working fluid. Our results provide strong evidence for the generality of the path integral framework for nonequilibrium response and a microscopic picture of the mobility response. Future work will focus on the development of effective hydrodynamic descriptions of the underlying fluid and tracer mobility.

A. R. P. and D. T. L. acknowledge helpful discussions with Kranthi Mandadapu and Cory Hargus. This work has been supported by NSF Grant No. CHE-1954580.

A. R. P. was also supported by the Heising-Simons Fellowship from the Kavli Energy Nanoscience Institute at UC Berkeley and D. T. L. acknowledges support from the Alfred P. Sloan Foundation.

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