Time-reversal symmetry breaking in two-dimensional nonequilibrium viscous fluids

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We study the rheological signatures of departure from equilibrium in two-dimensional viscous fluids with and without internal spin. Under the assumption of isotropy, we provide the most general linear constitutive relations for stress and couple stress in terms of the velocity and spin fields. Invoking Onsager's regression hypothesis for fluctuations about steady states, we derive the Green-Kubo formulas relating the transport coefficients to time-correlation functions of the fluctuating stress. In doing so, we show that one of the nonequilibrium transport coefficients, the odd viscosity, requires time-reversal symmetry breaking in the case of systems without internal spin. However, the Green-Kubo relations for systems with internal spin also show that there is a possibility for nonvanishing odd viscosity even when time-reversal symmetry is preserved. Furthermore, we find that breakdown of equipartition in nonequilibrium steady states results in the decoupling of the two rotational viscosities relating the vorticity and the internal spin.

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I. INTRODUCTION

This paper presents the consequences of time-reversal symmetry breaking at the microscale, and other signatures of nonequilibrium on emergent transport coefficients in two-dimensional viscous fluids. A motivation for this work is the recent emergence of the field of active matter, which studies systems that consume and dissipate energy at the particle scale. Active systems have been found to yield novel phase behavior [1–6] and continuum descriptions with unusual transport behavior [7–18], including odd viscosity [19,20]. They also provide insights into activity-mediated biological processes, including flows in the actin cortex [21,22] and collective motion in swarming and growing bacterial colonies [23–25].

Active systems with nonconservative forces at the microscale support nonequilibrium steady states different in nature from those arising from spatial gradients in temperature, pressure, or chemical potential by means of boundary conditions. The latter class of problems has been of intense interest for over a century and may be addressed within a well-established nonequilibrium thermodynamics formalism that unifies a variety of transport processes, building on the seminal work of Onsager, Prigogine, deGroot, and Mazur [26–29]. This approach is based on the local equilibrium hypothesis, expressing entropy production as a bilinear form of generalized thermodynamic forces X_α and fluxes J_α, with α enumerating the concerned transport process. The fluxes and forces are then taken to be related by linear laws,

$$J_\alpha = \sum_\beta L_{\alpha\beta} X_\beta. \tag{1}$$

The proportionality constants $L_{\alpha\beta}$, also referred to as transport coefficients, obey the celebrated Onsager reciprocal relations, $L_{\alpha\beta} = L_{\beta\alpha}$, derived by Onsager via invocation of the principle of microscopic time-reversibility (or time-reversal symmetry), and a regression hypothesis connecting the macroscopic boundary-driven gradient phenomena to fluctuations in equilibrium systems [26,27]. These same assumptions were used by Kubo, Yokota, and Nakajima to derive another set of prominent relations, the Green-Kubo relations, relating the constants $L_{\alpha\beta}$ to integrals of the time-correlation functions of the fluxes J_α in equilibrium systems [30,31].

Active matter systems, which break time-reversal symmetry at the microscale, still lack a unifying thermodynamic description for explaining emergent transport phenomena. In this work, we study the nonequilibrium viscous transport behavior of generic isotropic active systems in two dimensions with and without internal spin, investigate fluctuations in the nonequilibrium steady state, analyze the consequences of time-reversal symmetry breaking on the emergent transport coefficients, and demonstrate the breakdown of Onsager's reciprocal relations by deriving the Green-Kubo relations. In particular we elucidate the connection between time-reversal symmetry breaking and the emergence of a nonequilibrium transport coefficient, the odd viscosity. Furthermore, we show that breaking of equipartition leads to a decoupling of transport coefficients previously assumed to be related. We thus provide a first step towards developing a nonequilibrium thermodynamics formalism for transport phenomena in active media.

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II. ODD VISCOSITY AND TIME REVERSAL

We begin our treatment of emergent behavior in viscous nonequilibrium fluids with a brief discussion of odd viscosity, recently proposed as a consequence of time-reversal symmetry breaking in active media [19,32–34]. In a general theory of viscous fluids, a viscosity tensor η_{ijkl} defines a linear relation between the stress tensor T_{ij} and the velocity gradient $v_{k,l}$, where $(\cdot)_{,i}$ indicates the spatial partial derivative. Certain symmetries of the viscosity tensor are physically meaningful. It is well known that the symmetry $\eta_{ijkl} = \eta_{jikl}$ enforces the symmetry of the stress tensor, while the symmetry $\eta_{ijkl} = \eta_{ijlk}$ expresses its objectivity, i.e., its insensitivity to the antisymmetric part of the velocity gradient reflecting the rigid-body rotation of the fluid. Another symmetry that has recently attracted interest is $\eta_{ijkl} = \eta_{klij}$ [19,33]. Components of the viscosity that are antisymmetric with respect to this permutation do not contribute to the stress power $T_{ij}v_{i,j} = \eta_{ijkl}v_{i,j}v_{k,l}$, and are referred to as odd viscosities. These have interesting hydrodynamical consequences such as transverse response to shear strain that have been explored in both classical [19,34] and quantum [32] fluids. In the quantum setting, the odd viscosity is expected to appear in quantum Hall fluids [33].

In previous work on odd viscosity [19,33], the symmetry $\eta_{ijkl} = \eta_{klij}$ has been claimed as a necessary consequence of time-reversal symmetry on the basis of Onsager's reciprocal relations $L_{\alpha\beta} = L_{\beta\alpha}$. At first glance, it is plausible to take the symmetry $\eta_{ijkl} = \eta_{klij}$ of the viscosity to be a particular instance of Onsager reciprocity, identifying $\alpha = ij$ and $\beta = kl$, with stress being the flux of momentum and velocity gradient as the generalized thermodynamic force. This analogy is incorrect, as the reciprocal relations were developed for coupled thermodynamical transport processes using the entropy as a central tool [26,27]. The center-of-mass momentum of a fluid parcel is not a thermodynamic quantity and does not enter into any proper account of the entropy of that parcel. Thus an independent demonstration is required to prove that time-reversal symmetry breaking is necessary for the observation of odd viscosity and therefore the breakdown of the reciprocal relations for the viscosity tensor. This is one of the results we provide here, still using Onsager's particular insight, the regression hypothesis connecting the fluctuations in the steady state to macroscopic boundary-driven gradients in velocity.

III. CONSERVATION LAWS

In establishing the breakdown of reciprocal relations in active fluids, we study generalized viscous fluids sustaining internal spin [35–37]. These are relevant to systems ranging from chiral active and granular materials [37,38] to biological systems behaving as active gels [39,40], while exhibiting interesting behavior due to active driving forces, namely, edge flows and topological localization [18]. To study two-dimensional fluids with internal structure, we take as fundamental dynamical fields the velocity vector v_i and the scalar internal spin m. Conservation of linear and angular momentum is guaranteed by the balance equations

$$\rho\dot{v}_i = T_{ij,j}, \tag{2}$$

$$\rho\dot{m} = C_{i,i} - \epsilon_{ij}T_{ij}, \tag{3}$$

with T_{ij} being the stress tensor and C_i being the couple stress or spin flux. The dot indicates the convective or material derivative $\partial_t + v_i\partial_i$, and ϵ_{ij} is the two-dimensional Levi-Civita tensor. Such a microstructural continuum theory was proposed by Dahler and Scriven [35,36]. The coupling term $-\epsilon_{ij}T_{ij}$ preserves conservation of total angular momentum while permitting the existence of an antisymmetric component of stress. The hydrodynamic equations (2) and (3) are shown to arise for a class of active systems consisting of dumbbell particles subjected to active or nonconservative rotary forces [16,41].

IV. CONSTITUTIVE RELATIONS AND ISOTROPY

To close Eqs. (2) and (3) for v_i and m, we require constitutive equations relating the stress T_{ij} and couple stress C_i to the fields v_i and m. We assume that these relations are linear, Galilean invariant, and contain derivatives of the fields only up to first order. The most general linear constitutive relations are then given by

$$T_{ij} = \eta_{ijkl}v_{k,l} + \gamma_{ij}m + \xi_{ijk}m_{,k}, \tag{4}$$

$$C_i = \beta_{ijk}v_{j,k} + \kappa_i m + \alpha_{ij}m_{,j}, \tag{5}$$

where repeated indices are summed, and $\boldsymbol{\eta}$, $\boldsymbol{\gamma}$, $\boldsymbol{\xi}$, $\boldsymbol{\beta}$, $\boldsymbol{\kappa}$, and $\boldsymbol{\alpha}$ are linear maps.

Imposing isotropy further restricts the couplings in Eqs. (4) and (5). Isotropic tensors of any rank in dimension n may be expressed as linear combinations of terms consisting only of the rank-two Kronecker tensor δ_{ij} and the rank-n Levi-Civita tensor $\epsilon_{i_1,\ldots,i_n}$ (see Sec. I in the Supplemental Material (SM) [42]). In two dimensions, both of these are rank two, so there are no nonzero isotropic tensors of odd rank. This forbids the existence of nontrivial isotropic linear maps between tensors with ranks differing by an odd number. For instance, the couple stress C_i, a vector, cannot depend on the spin density m, a scalar, or the velocity gradient $v_{i,j}$, a rank-two tensor. Similarly, the stress tensor T_{ij}, a rank-two tensor, cannot depend on the spin gradient $m_{,i}$, a vector. Therefore, the most general isotropic constitutive equations have the form

$$T_{ij} = \eta_{ijkl}v_{k,l} + \gamma_{ij}m, \quad C_i = \alpha_{ij}m_{,j}. \tag{6}$$

The maps γ_{ij} and α_{ij} may be expressed as

$$\gamma_{ij} = \gamma_1\delta_{ij} + \gamma_2\epsilon_{ij}, \quad \alpha_{ij} = \alpha_1\delta_{ij} + \alpha_2\epsilon_{ij}. \tag{7}$$

The viscosity tensor η_{ijkl} is an element of the six-dimensional space of isotropic rank-four tensors in two dimensions (see Sec. I in the SM [42]). An orthogonal basis $\mathbf{s}^{(\alpha)}$ for this space is provided in Table I, along with the symmetry properties of the basis elements under various index permutations of physical significance. We can express the viscosity tensor as a linear combination of these basis elements:

$$\eta_{ijkl} = \sum_{\alpha=1}^{6}\lambda_\alpha s^{(\alpha)}_{ijkl}. \tag{8}$$

In Table I, we provide the components $s^{(\alpha)}_{ijkl}v_{k,l}$ of the stress tensor due to each of the basis tensors, elucidating the physical meaning of each coefficient. The bulk viscosity λ_1 and shear viscosity λ_2 resist compression and shearing as in a typical

TABLE I. The tensors $s^{(\alpha)}_{ijkl}$ form a basis for the isotropic rank-four tensors in two dimensions, and are orthogonal with respect to the inner product $A_{ijkl}B_{ijkl}$. This basis has been chosen to be an eigenbasis for the index permutations $i \leftrightarrow j$ and $k \leftrightarrow l$, corresponding to the symmetry and objectivity of the stress tensor, respectively. It is also an eigenbasis for the mirror transformation $x_1 \mapsto -x_1$, $x_2 \mapsto x_2$, also known as the parity transformation (P). Four of these basis tensors are also eigenvectors of the index permutation $i \leftrightarrow k$ and $j \leftrightarrow l$, and we also indicate the parity of the basis tensors under this transformation. In the last column, we provide the component of the stress $T_{ij} = \eta_{ijkl}v_{k,l}$ due to each basis element of the viscosity. The symmetric traceless velocity gradient is defined as $\mathring{u}_{ij} = \frac{1}{2}(v_{i,j} + v_{j,i} - v_{k,k}\delta_{ij})$, and the vorticity as $\omega = -\frac{1}{2}\epsilon_{ij}v_{i,j}$. We also use the matrices $\boldsymbol{\sigma}_z = [1, 0; 0, -1]$ and $\boldsymbol{\sigma}_x = [0, 1; 1, 0]$, which are basis elements of the pure shear modes of the velocity gradient that transform into each other under rotation. The tensor $\boldsymbol{\sigma}_z \otimes \boldsymbol{\sigma}_x$ maps a pure shear mode of the velocity gradient to a rotated pure shear mode of the stress. A complete eigenbasis $\boldsymbol{e}^{(\beta)}$ for the permutation $ij \leftrightarrow kl$ is presented in Sec. I of the SM [42].

Basis tensor	Components	$i \leftrightarrow j$	$k \leftrightarrow l$	$ij \leftrightarrow kl$	P	$s^{(\alpha)}_{ijkl}v_{k,l}$
$s^{(1)}_{ijkl}$	$\delta_{ij}\delta_{kl}$	+	+	+	+	$(\nabla \cdot \mathbf{v})\delta_{ij}$
$s^{(2)}_{ijkl}$	$\delta_{ik}\delta_{j\ell} + \delta_{i\ell}\delta_{jk} - \delta_{ij}\delta_{kl}$	+	+	+	+	$2\mathring{\mathbf{u}}$
$s^{(3)}_{ijkl}$	$\epsilon_{ij}\epsilon_{kl}$	−	−	+	+	$-2\omega\epsilon_{ij}$
$s^{(4)}_{ijkl}$	$\epsilon_{ik}\delta_{j\ell} + \epsilon_{j\ell}\delta_{ik}$	+	+	−	−	$(\boldsymbol{\sigma}_z \otimes \boldsymbol{\sigma}_x - \boldsymbol{\sigma}_x \otimes \boldsymbol{\sigma}_z) : \mathring{\mathbf{u}}$
$s^{(5)}_{ijkl}$	$\epsilon_{ik}\delta_{j\ell} - \epsilon_{j\ell}\delta_{ik} + \epsilon_{ij}\delta_{k\ell} + \epsilon_{k\ell}\delta_{ij}$	−	+	N/A	−	$(\nabla \cdot \mathbf{v})\epsilon_{ij}$
$s^{(6)}_{ijkl}$	$\epsilon_{ik}\delta_{j\ell} - \epsilon_{j\ell}\delta_{ik} - \epsilon_{ij}\delta_{k\ell} - \epsilon_{k\ell}\delta_{ij}$	+	−	N/A	−	$4\omega\delta_{ij}$

Newtonian fluid. The rotational viscosity λ_3 resists rotation, corresponding to the appearance of a torque in response to nonvanishing vorticity, breaking both symmetry and objectivity of the stress tensor. All three of these components of the viscosity are even under mirror symmetry, implying that they may arise in nonchiral systems.

The other three components of the viscosity are odd under mirror symmetry and thus should be expected to vanish in nonchiral systems. The odd viscosity λ_4, corresponding to a term that violates the permutation symmetry $\eta_{ijkl} = \eta_{klij}$, responds to pure shear along one axis with pure shear stress along an axis rotated by $\pi/4$. Equivalently, it responds to simple shear along one axis with pressure or tension along the orthogonal axis, depending on the sign of the shear. An interesting feature of this component of the viscosity is that it is nondissipative in the sense that it does not contribute to the stress power $T_{ij}v_{i,j}$. This term satisfies both objectivity and symmetry of the stress tensor, so that it is compatible with conservation of angular momentum even in the absence of a mechanism coupling internal spin to the velocity gradient.

Finally, the component λ_5 responds to compression with torque, breaking symmetry of the stress, while the component λ_6 responds to vorticity with isotropic pressure, breaking objectivity. The corresponding basis tensors $\mathbf{s}^{(5)}$ and $\mathbf{s}^{(6)}$ both violate the symmetry $\eta_{ijkl} = \eta_{klij}$. They span a two-dimensional subspace with one even and one odd direction under this index permutation, so that there are in fact two independent odd components of the viscosity.

V. ONSAGER'S REGRESSION HYPOTHESIS AND GREEN-KUBO RELATIONS

The reciprocal relations, or symmetry relations of the transport coefficients were derived by Onsager in a seminal work connecting macroscopic phenomena and transport coefficients to time correlations of fluctuations of related variables at the microscopic level, using regression hypothesis for the decay of fluctuations and the principle of microscopic reversibility (or time-reversal symmetry) [26,27]. This connection between microscopic reversibility and the symmetry of macroscopic transport coefficients may also be established via the Green-Kubo relations [31], which provide explicit microscopic expressions for the transport coefficients. Note that Onsager's regression hypothesis and time-reversal symmetry are independent assumptions, and one may be invoked without the other. In what follows, we invoke only the regression hypothesis for decay of fluctuations of in nonequilibrium steady states and determine the effects of time-reversal symmetry breaking on transport coefficients. In particular, we derive the Green-Kubo formulas relating the viscous coefficients introduced in Eqs. (7) and (8) to the stress-stress time-correlation function in a fluctuating steady state, starting from an assumption on the fluctuations in the spirit of Onsager's regression hypothesis. The philosophy adopted here is to suppose that the fields v_i, m, T_{ij}, and C_i are fluctuating or stochastic rather than deterministic, but that small fluctuations about a steady state behave, in expectation, in the same manner as the deterministic transport equations would predict. In other words, a viscous fluid is best described in different regimes by either a deterministic or a stochastic theory, and the two must be related in some plausible way. This is the informal content of Onsager's regression hypothesis [26,27,30,31].

We now provide a more formal presentation of the statement of the regression hypothesis in a general setting, which will be central to the derivation of Green-Kubo relations. Suppose a system is characterized by some set of complex variables A_i and B_j, and that the system is described by a deterministic theory that obeys the linear dynamical (or conservation) and constitutive equations

$$\frac{dA_i}{dt} = \sum_j M_{ij}B_j, B_j = \sum_i S_{ji}A_i, \tag{9}$$

where M_{ij} and S_{kl} are constant coefficients, and $A_i = B_j = 0$ is a stable fixed point. These lead to the transport equations

$$\frac{dA_i}{dt} = \sum_{j,k} M_{ij}S_{jk}A_k, \tag{10}$$

where any external perturbation to the variables A_i decays with a characteristic relaxation time $\tau_{\rm r} \approx \frac{1}{M_{ij}S_{jk}}$.

Extending Onsager's regression hypothesis to nonequilibrium steady states [27], we suppose that spontaneous fluctuations about the steady state decay according to the transport equation (10) in expectation, in the sense that

$$\frac{\langle A_i(t+\Delta t)\rangle_{t,\mathbf{a}} - a_i}{\Delta t} = \sum_{j,k} M_{ij} S_{jk} a_k, \tag{11}$$

where the subscript indicates that the expectation is taken over the subensemble of trajectories satisfying $A_i(t) = a_i$. Note that we have in mind steady states of active matter systems subjected to microscopic nonconservative forces rather than those arising from spatial gradients due to boundary conditions. In Eq. (11), Δt is chosen to be small in comparison with the macroscopic relaxation time τ_r, but sufficiently large compared with the microscopic or molecular timescales [27,31]. This is the mathematical statement of the regression hypothesis.

Following Kubo-Yokota-Nakajima [31], we may derive from the regression (11) (see Sec. III in the SM [42]) the generalized Green-Kubo relations

$$M_{ij} S_{jk} \langle A_k(0) A_r^*(0)\rangle = -\int_0^\infty \langle \dot{A}_i(t) \dot{A}_r^*(0)\rangle dt \tag{12}$$

$$= -M_{ij} M_{rk}^* \int_0^\infty \langle B_j(t) B_k^*(0)\rangle dt, \tag{13}$$

where the averages are taken over the steady-state ensemble of trajectories, $(\cdot)^*$ denotes complex conjugation, and repeated indices are summed. Deriving Eq. (12) requires an important condition on the separation of timescales:

$$\tau_{\text{corr}} \ll \Delta t \ll \frac{1}{M_{ij} S_{jk}}, \tag{14}$$

where τ_{corr} is the timescale associated with the decay of the correlation functions $\langle \dot{A}_j(t) \dot{A}_r^*(0)\rangle$.

VI. GREEN-KUBO RELATIONS FOR VISCOUS FLUIDS

For our system of nonequilibrium fluids, the role of the variables A_i and B_j will be played by the large-wavelength components of the fluctuations of the fields v_i, m, T_{ij}, and C_i about the steady state with $v_i = 0$ and $m = \text{const}$. The evolution of these components is governed by the linearized Fourier forms of the linear- and angular-momentum balance equations (see Sec. II in the SM [42]). Invoking a regression hypothesis on these variables in the spirit of Eq. (11) and examining the large-wavelength limit of the fluctuations yields the following Green-Kubo relations for the transport coefficients (see Sec. V in the SM [42] for detailed derivations):

$$\gamma_1 = \frac{1}{2\rho_0 \nu} \delta_{ij} \epsilon_{kl} \mathcal{T}^{ijkl}, \tag{15}$$

$$\gamma_2 = \frac{1}{2\rho_0 \nu} \epsilon_{ij} \epsilon_{kl} \mathcal{T}^{ijkl}, \tag{16}$$

$$\lambda_1 + 2\lambda_2 + \lambda_3 - \frac{\gamma_1 \pi}{2\mu} + \frac{\gamma_2 \tau}{2\mu} = \frac{1}{2\rho_0 \mu} \delta_{ik} \delta_{jl} \mathcal{T}^{ijkl}, \tag{17}$$

$$\lambda_4 + \lambda_5 + \lambda_6 - \frac{\gamma_1 \tau}{4\mu} - \frac{\gamma_2 \pi}{4\mu} = \frac{1}{4\rho_0 \mu} \epsilon_{ik} \delta_{jl} \mathcal{T}^{ijkl}, \tag{18}$$

$$\lambda_5 - \frac{\gamma_2 \pi}{4\mu} = \frac{1}{8\rho_0 \mu} \epsilon_{ij} \delta_{kl} \mathcal{T}^{ijkl}, \tag{19}$$

$$\lambda_3 + \frac{\gamma_2 \tau}{2\mu} = \frac{1}{4\rho_0 \mu} \epsilon_{ij} \epsilon_{kl} \mathcal{T}^{ijkl}, \tag{20}$$

where $\mathcal{T}^{ijkl}$ is the time-integrated stress-stress correlator

$$\mathcal{T}^{ijkl} = \frac{1}{L^4} \int_0^\infty dt \int d^2\mathbf{x}\, d^2\mathbf{y} \langle \delta T_{ij}(\mathbf{x}, t) \delta T_{kl}(\mathbf{y}, 0)\rangle, \tag{21}$$

and μ, ν, τ, and π are the steady-state correlation functions defined by

$$\mu \delta_{ij} = \frac{1}{L^4} \int \langle \delta v^i(\mathbf{x}) \delta v^j(\mathbf{y})\rangle d^2\mathbf{x}\, d^2\mathbf{y}, \tag{22}$$

$$\pi = \frac{1}{L^4} \int (y^i - x^i) \langle \delta v^i(\mathbf{x}) \delta m(\mathbf{y})\rangle d^2\mathbf{x}\, d^2\mathbf{y}, \tag{23}$$

$$\tau = \frac{1}{L^4} \int \epsilon_{kr} (y^r - x^r) \langle \delta m(\mathbf{x}) \delta v^k(\mathbf{y})\rangle d^2\mathbf{x}\, d^2\mathbf{y}, \tag{24}$$

$$\nu = \frac{1}{L^4} \int \langle \delta m(\mathbf{x}) \delta m(\mathbf{y})\rangle d^2\mathbf{x}\, d^2\mathbf{y}. \tag{25}$$

In Eqs. (15)–(25), δa indicates the fluctuation about the steady-state value of a.

The constants μ and ν provide an estimate of the effective kinetic temperature in the steady state and by the equipartition theorem are proportional to the Boltzmann temperature in the special case of equilibrium systems. The constant τ measures the correlation of the internal spin with the fluid vorticity, in other words, the correlation between the internal and external angular-momentum density fields. The constant π measures the correlation of the internal spin with the fluctuating divergence of the velocity field.

Several features of the Green-Kubo relations (15)–(20) are noteworthy. In the absence of internal spin (or the absence of a mechanism for coupling internal spin to the velocity field), $\gamma_1 = \gamma_2 = 0$ by assumption and $\lambda_3 = \lambda_5 = 0$ by conservation of angular momentum. Then we are left with the Green-Kubo relations

$$\lambda_1 + 2\lambda_2 = \frac{1}{2\rho_0 \mu} \delta_{ik} \delta_{jl} \mathcal{T}^{ijkl}, \tag{26}$$

$$\lambda_4 + \lambda_6 = \frac{1}{4\rho_0 \mu} \epsilon_{ik} \delta_{jl} \mathcal{T}^{ijkl}. \tag{27}$$

If we assume that the stress tensor is objective, then $\lambda_6 = 0$ and we are left with a Green-Kubo relation for the odd viscosity λ_4 given by

$$\begin{aligned}\lambda_4 = \frac{1}{4\rho_0 \mu} \frac{1}{L^4} \int_0^\infty dt \int d^2\mathbf{x}\, d^2\mathbf{y} \\ \times [\langle \delta T_{11}(\mathbf{x}, t) \delta T_{21}(\mathbf{y}, 0)\rangle - \langle \delta T_{21}(\mathbf{x}, t) \delta T_{11}(\mathbf{y}, 0)\rangle \\ + \langle \delta T_{12}(\mathbf{x}, t) \delta T_{22}(\mathbf{y}, 0)\rangle - \langle \delta T_{22}(\mathbf{x}, t) \delta T_{12}(\mathbf{y}, 0)\rangle].\end{aligned} \tag{28}$$

It is clear from Eq. (28) that only the component of the stress autocorrelation function that is odd under time reversal survives, thus demonstrating that nonvanishing odd viscosity $\lambda_4 \neq 0$ requires breaking time-reversal symmetry at the level of the steady-state stress fluctuations for fluids without internal spin.

Now allowing for coupling of internal spin to the fluid velocity, we may observe from Eqs. (16) and (20) that

$$2\lambda_3 = \left(\frac{\nu - \tau}{\mu}\right) \gamma_2. \tag{29}$$

In an equilibrium system, $\nu = \mu$ by equipartition and there exist no correlations between internal spin and vorticity, so that $\tau = 0$. Then $\gamma_2 = 2\lambda_3$, so that there is a single parameter characterizing the response of the stress to both the spin m and the vorticity ω. This feature is assumed in many previous works on out-of-equilibrium active systems [19,20,38–40]. It should be noted that such active systems may break equipartition in the steady state so that in general $\nu - \tau \neq \mu$, which leads to decoupling of the two rotational viscosity coefficients coupling the vorticity and internal spin, and therefore this assumption must be revisited.

Finally, we note that, in a system with internal spin that obeys time-reversal symmetry at the level of the stress correlations, the Green-Kubo relation (18) involving the odd viscosity reduces to

$$\lambda_4 + \lambda_5 + \lambda_6 = \frac{\gamma_1 \tau}{4\mu} + \frac{\gamma_2 \pi}{4\mu}. \qquad (30)$$

Thus λ_4 need not necessarily vanish. Therefore, it is possible that there are systems that do not break time-reversal symmetry at the level of stress correlations, yet do exhibit odd viscosity due to a coupling of internal spin to fluid velocity. This possibility merits future consideration.

VII. CONCLUSION

In this work, we have made progress towards the goal of understanding transport phenomena in systems that break time-reversal symmetry. By deriving Green-Kubo formulas via an Onsager regression hypothesis, we have put on stronger footing the claim that, in systems without internal spin, nonvanishing odd viscosity requires breaking time-reversal symmetry at the level of stress-stress correlations. However, in systems with internal spin, we cannot rule out the possibility of nonvanishing odd viscosity even when this symmetry is preserved. Furthermore, we have demonstrated that breaking of equipartition leads to modification of the coupling between internal spin and fluid vorticity. Finally, we note that our Green-Kubo formulas for shear and odd viscosities with an effective temperature have been validated in a model active system consisting of dumbbells subjected to active rotary forces, by comparison to independent measurements obtained from nonequilibrium flow studies [41]. In particular, we show that nonvanishing odd viscosity arises only under the application of active forces and corresponds to breaking of time-reversal symmetry of stress correlation functions as predicted in this work.

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