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Onsager's Variational Principle for Nonreciprocal Systems with Odd Elasticity

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Using Onsager's variational principle, we derive dynamical equations for a nonequilibrium active system with odd elasticity. The elimination of the extra variable that is coupled to the nonequilibrium driving force leads to the nonreciprocal set of equations for the material coordinates. The obtained nonreciprocal equations manifest the physical origin of the odd elastic moduli that are proportional to the nonequilibrium force and the friction coefficients. Our approach offers a systematic and consistent way to derive nonreciprocal equations for active matter in which the time-reversal symmetry is broken.

Active matter is composed of self-driven units that consume energy from the environment and drive the system into out-of-equilibrium situations.^{1,2)} The motion of active particles is typically directional because the activity and local orientation lead to the broken time-reversal symmetry.³⁾ A practical way to describe the collective behaviors of active particles is to write down equations for slow variables by considering all possible hydrodynamic force and flux pairs.⁴⁻⁶⁾

Concerning the dynamics of passive systems, it is known that Onsager's variational principle (OVP) provides us with proper dynamical equations that automatically satisfy Onsager's reciprocal relations and the second law of thermodynamics. In this formulation, dynamical equations are obtained by minimizing a Rayleighian that is the sum of dissipation function and time derivative of free energy. OVP has been proven powerful for various passive dynamical problems in soft matter such as polymer systems, 100 membranes, 111 and liquid droplets. Nevertheless, OVP has not yet been employed for active systems due to the difficulties in determining the free energy or the dissipation function under nonequilibrium conditions. One attempt is to introduce an additional term in Rayleighian accounting for the work power done by the active forces. 13,14)

In this letter, we focus on the new concept of active systems, i.e., odd elasticity introduced by Scheibner et al. ^{15,16)} Odd elasticity arises from an anti-symmetric (odd) component of the elastic modulus tensor. It violates the energy conservation law and thus can exist only in active materials. Unlike passive materials, a finite amount of work can be extracted in odd elastic systems through quasi-static cycle of deformations. ^{15,16)} Recently, the present authors proposed a thermally driven microswimmer with odd elasticity and demonstrated that it can exhibit a directional locomotion. ^{17,18)} We also showed that anti-symmetric parts of the time-correlation functions in odd Langevin systems are proportional to the odd elasticity. ¹⁹⁾ Moreover, the concept of odd elasticity can be extended to quantify the nonreciprocality of active micromachines such as enzymes and motor proteins. ^{20,21)}

The purpose of our work is twofold. First, we show that typical dynamical equations of an active system can be obtained within the framework of OVP, which has a different structure compared with the previous approach. ^{13,14)} In

addition to the regular material coordinates, we introduce an extra coordinate that is conjugate to the nonequilibrium active force (or torque) in the free energy. Being constantly driven by the active force, this extra coordinate transmits energy to the material coordinates through dissipative processes. Importantly, we obtain nonreciprocal (or nonconservative²²) dynamical equations for the material coordinates after eliminating the extra coordinate from the coupled equations derived from OVP.

Second, the obtained nonreciprocal (coarse-grained) equations for the material coordinates provide us with the physical origin of the odd elasticity that was previously introduced merely by symmetry consideration. ^{15,16} We explicitly show that the odd elastic moduli are proportional to the nonequilibrium active force and the friction coefficients appearing in the dissipation function of OVP. We demonstrate our argument first by using a simplified model with only two material degrees of freedom and then later generalize it to a continuum model. Although we limit our discussion only to odd elastic systems, our approach of using OVP and nonequilibrium extra variable offers a systematic way to obtain nonreciprocal equations for other active systems.

To demonstrate the idea of applying OVP for active systems, we consider a two-dimensional (2D) system with a particle confined by four springs of stiffness k and located above a rotary plate in a viscous environment, as shown in Fig. 1. In this overdamped situation, the mass of the particle can be neglected. The disk rotates with an angular velocity $\dot{s} = ds/dt$ (s being the angle of rotation) due to a constant driving torque f exerted on the disk. Notice that f can take both positive and negative values and nonzero f breaks the time-reversal symmetry. When the particle moves from the origin by a displacement $\mathbf{u} = (u_x, u_y)$, the total potential energy U and its time derivative can be written as

$$U = \frac{k}{2} (u_x^2 + u_y^2) - fs, \tag{1}$$

$$\dot{U} = k(u_x \dot{u}_x + u_y \dot{u}_y) - f\dot{s}. \tag{2}$$

Because the confined particle is located above and close to the rotary disk, linear viscous friction arises from the relative motion between the particle and the disk. (One can also assume a lubricating layer between the disk and the particle.) Then the associated dissipation function Φ is written as

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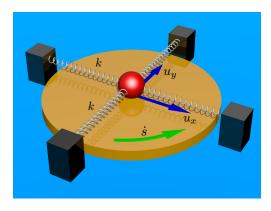


Fig. 1. (Color online) A particle confined by the four springs of stiffness k and moving above a disk rotating with an angular velocity \dot{s} . The displacements of the particle are denoted by u_x and u_y . The whole setup is immersed in a viscous fluid that causes dissipation.

$$\Phi = \frac{\zeta_u}{2} (\dot{u}_x^2 + \dot{u}_y^2) + \frac{\zeta_s}{2} \dot{s}^2 + \frac{\xi}{2} (u_x \dot{s} - \dot{u}_y)^2 + \frac{\xi}{2} (u_y \dot{s} + \dot{u}_x)^2,$$
(3)

where ζ_u , ζ_s , and ξ are the positive friction coefficients corresponding to the particle velocity, the rotational velocity of the disk, and the relative motion between them, respectively. The last two terms in Eq. (3) arise when the cross product of $(0,0,\dot{s})$ and $(u_x,u_y,0)$ differs from $(\dot{u}_x,\dot{u}_y,0)$. Importantly, these terms indicate that finite \dot{s} induces the quantities \dot{u}_y/u_x and \dot{u}_x/u_y in an anti-symmetric way. This is the key idea that would later lead to the nonreciprocal interaction of the system, which is a common feature of active systems. ^{19,22)} In general, the two coefficients ξ can be different, but we consider the simplest case here.

The Rayleighian of the system is given by the sum of Eqs. (2) and (3), i.e., $R = \dot{U} + \Phi^{.8,9)}$ Minimization of R with respect to \dot{u}_x , \dot{u}_y , and \dot{s} leads to the following set of coupled equations:

$$\zeta_u \dot{u}_x + k u_x + \xi(u_v \dot{s} + \dot{u}_x) = 0, \tag{4}$$

$$\zeta_u \dot{u}_v + k u_v - \xi(u_x \dot{s} - \dot{u}_v) = 0, \tag{5}$$

$$\zeta_{s}\dot{s} + \xi(u^{2}\dot{s} - u_{x}\dot{u}_{y} + \dot{u}_{x}u_{y}) = f, \tag{6}$$

where $u^2 = u_x^2 + u_y^2$ in Eq. (6). Equations (4) and (5) describe the motion of the particle experiencing both the frictional and elastic forces, while the terms with ξ express how the particle velocity couples with the external chiral motion \dot{s} . On the other hand, Eq. (6) means that the angular momentum $u_x\dot{u}_y - u_y\dot{u}_x$ is generated by the driving torque f through the friction ξ .

Next, we use Eq. (6) to eliminate the variable \dot{s} from Eqs. (4) and (5). Then the nonlinear dynamics of u_x and u_y can be expressed in the matrix form as

$$\Gamma \begin{pmatrix} \dot{u}_x \\ \dot{u}_y \end{pmatrix} = \mathbf{E} \begin{pmatrix} u_x \\ u_y \end{pmatrix},\tag{7}$$

where the friction matrix Γ and the generalized elastic matrix E are introduced by

$$\Gamma = \begin{pmatrix} \zeta_{u} + \frac{\xi(\xi u_{x}^{2} + \zeta_{s})}{\xi u^{2} + \zeta_{s}} & \frac{\xi^{2} u_{x} u_{y}}{\xi u^{2} + \zeta_{s}} \\ \frac{\xi^{2} u_{x} u_{y}}{\xi u^{2} + \zeta_{s}} & \zeta_{u} + \frac{\xi(\xi u_{y}^{2} + \zeta_{s})}{\xi u^{2} + \zeta_{s}} \end{pmatrix}, \tag{8}$$

$$\mathbf{E} = \begin{pmatrix} -k & -\frac{f\xi}{\xi u^2 + \zeta_s} \\ \frac{f\xi}{\xi u^2 + \zeta_s} & -k \end{pmatrix}. \tag{9}$$

Notice that Γ is symmetric and positive-definite while \mathbf{E} is anti-symmetric. Hence, the elimination of \dot{s} gives rise to the nonreciprocal interaction between u_x and u_y .^{17,19)} We emphasize that the anti-symmetric off-diagonal components of \mathbf{E} are present only when $f \neq 0$ corresponding to the nonequilibrium situation. Moreover, one can easily show that $(u_x, u_y) = (0, 0)$ is the unique equilibrium point that is globally stable.

For the large-displacement limit, $\xi u^2 \gg \zeta_s$, the above friction and elastic matrices become

$$\Gamma \approx \begin{pmatrix} \zeta_u + \xi u_x^2 / u^2 & \xi u_x u_y / u^2 \\ \xi u_x u_y / u^2 & \zeta_u + \xi u_y^2 / u^2 \end{pmatrix}, \tag{10}$$

$$\mathbf{E} \approx \begin{pmatrix} -k & -f/u^2 \\ f/u^2 & -k \end{pmatrix}. \tag{11}$$

The off-diagonal elements of E become smaller when the displacement amplitude u^2 is larger.

On the other hand, for the small-displacement limit, $\xi u^2 \ll \zeta_s$, Eq. (7) can be simply linearized as

$$(\zeta_u + \xi) \begin{pmatrix} \dot{u}_x \\ \dot{u}_y \end{pmatrix} = \begin{pmatrix} -k & -f\xi/\zeta_s \\ f\xi/\zeta_s & -k \end{pmatrix} \begin{pmatrix} u_x \\ u_y \end{pmatrix}. \tag{12}$$

The frictional factor $\zeta_u + \xi$ on the left-hand side indicates that the energy associated with u_x and u_y dissipates through the relative motion with respect to the environment and also to the rotary disk. The off-diagonal elements on the right-hand side imply that the degree of nonreciprocality is controlled by the ratio ξ/ζ_s . This ratio characterizes the relative energy injected to the displacement coordinates u_x and u_y with respect to that dissipated through the variable \dot{s} .

The linear equation in Eq. (12) can easily be solved as

$$u_x = U_0 e^{-\Lambda t} \sin(\Omega t - \phi), \quad u_y = U_0 e^{-\Lambda t} \cos(\Omega t - \phi), \quad (13)$$

where the decay rate Λ and the angular frequency Ω are given by $\Lambda = k/(\zeta_u + \xi)$ and $\Omega = f\xi/[(\zeta_u + \xi)\zeta_s]$, respectively. In the above, the amplitude U_0 and the phase difference ϕ are determined by the initial condition. The system relaxes to its steady state through the conservative restoring force provided by the stiffness k and the dissipation due to the total friction, $\zeta_u + \xi$. On the other hand, the oscillatory motion is driven by the torque source f weighted by the ratio ξ/ζ_s .

We choose the units for length and time as $(\zeta_s/\xi)^{1/2}$ and ζ_u/k , respectively. The other dimensionless parameters are ξ/ζ_u and $f\xi/(k\zeta_s) = \Omega/\Lambda$. In Fig. 2, we show the time evolution of u_x and u_y by numerically solving the nonlinear dynamics in Eq. (7) for certain parameters. We see that u_x and u_y do not oscillate when their amplitudes are large, while the oscillatory behavior becomes pronounced when the amplitudes become smaller. The phase difference between the oscillations of u_x and u_y are almost $\pi/2$. These behaviors are consistent with the behaviors for the large-amplitude [Eqs. (10) and (11)] and small-amplitude [Eq. (12)] cases.

Next, we shall extend the above simple model to a 2D continuum model with odd elasticity. Due to the translational symmetry, the free energy of an elastic material should be a

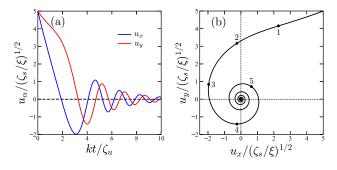


Fig. 2. (Color online) (a) Time evolution of dimensionless u_x (blue) and u_y (red) as a function of dimensionless time kt/ζ_u when the initial condition is $u_x/(\zeta_s/\xi)^{1/2} = u_y/(\zeta_s/\xi)^{1/2} = 5$ [see Eq. (7)]. The other parameters are $\xi/\zeta_u = 1.5$ and $f\xi/(k\zeta_s) = 10$. The oscillatory motion due to the nonreciprocal force starts to appear when u_x and u_y become smaller. (b) The parametric plot of (a) on the plane of u_x and u_y . The numbers on the trajectory indicate the dimensionless time.

functional of the gradients of displacement. We consider position dependent 2D displacement field $\mathbf{u}(\mathbf{r}) = (u_x(\mathbf{r}), u_y(\mathbf{r}))$ where $\mathbf{r} = (x, y)$. When the deformation is small, the free energy F can be expressed in terms of quadratic forms of nonsymmetrized strain tensor $u_{ij} = \partial u_i/\partial r_i = \partial_i u_i$ (i, j = x, y) as 9,23

$$F = \int d\mathbf{r} \left[\frac{K}{2} w^2 + \frac{G}{4} (u_{ij} + u_{ji} - w \delta_{ij})^2 - fs \right], \quad (14)$$

where the summation over repeated indices is assumed, $w = u_{ii}$ is the trace of the strain tensor, and δ_{ij} is the Kronecker delta. In the above, K and G are the standard bulk and shear moduli, respectively, and f is a constant nonequilibrium force conjugate to the extra variable $s(\mathbf{r})$ that is independent of the strain. For an odd elastic material, \dot{s} must be rotational (with the ability to drive a force field with nonzero curl in the material coordinates), breaking both the timereversal symmetry and the conservative nature of the even elasticity. For example, f is the torque density determined by odd microdevices such as internal actuators²⁴⁾ and s is the dimensionless microrotation field.²⁵⁾ The time derivative of F can be written as⁹⁾

$$\dot{F} = \int d\mathbf{r} [\sigma_{ij}\partial_j \dot{u}_i - f\dot{s}], \tag{15}$$

where $\sigma_{ij} = Kw\delta_{ij} + G(u_{ij} + u_{ji} - w\delta_{ij})$ is the elastic stress tensor

Next, we discuss the dissipation function of a 2D odd material by using the symmetry argument. To explain the nonreciprocal relations between strains and strain rates, we introduce the following four deformation bases $\tau_0 = u_{xx} + u_{yy}$, $\tau_1 = u_{yx} - u_{xy}$, $\tau_2 = u_{yy} - u_{xx}$, and $\tau_3 = u_{xy} + u_{yx}$ representing dilation, rotation, and two types of shear, respectively. By assuming that the whole system is isotropic, it has been shown that the elastic modulus tensor $C_{ijk\ell}$ can be written as 15,16)

$$C_{ijk\ell} = K\delta_{ij}\delta_{k\ell} - A\epsilon_{ij}\delta_{k\ell} - \Xi\delta_{ij}\epsilon_{k\ell} + \Upsilon\epsilon_{ij}\epsilon_{k\ell}$$

+ $G(\delta_{i\ell}\delta_{jk} + \delta_{ik}\delta_{j\ell} - \delta_{ij}\delta_{k\ell}) + K^{o}(\epsilon_{ik}\delta_{j\ell} + \epsilon_{j\ell}\delta_{ik}),$

where ϵ_{ij} is the 2D Levi-Civita tensor, and K and G are the aforementioned bulk and shear moduli, respectively. For simplicity, we further assume that the solid-body rotations do

not induce stress (called objectivity) and set $\Xi = \Upsilon = 0$. The modulus A converts dilation into torque $(\tau_0 \to \dot{\tau}_1)$, and K^o converts shear strains to shear stresses in an antisymmetric way $(\tau_3 \to \dot{\tau}_2 \text{ and } \tau_2 \to -\dot{\tau}_3)$. Both A and K^o correspond to the antisymmetric components of $C_{ijk\ell}$ under the exchange of $ij \leftrightarrow k\ell$ and they represent the odd elastic moduli. ^{15,16}

Adapting the above symmetry argument, we consider the dissipation mechanism such that strain rates are induced by strains through the rotational field \dot{s} . Notice that \dot{s} conjugates here to the strain because we consider a continuum model with internal actuators such as the one shown in Fig. 1. Among all the possible dissipation terms $(\tau_{\alpha}\dot{s}\pm\dot{\tau}_{\beta})^2$ $(\alpha,\beta=0,\ldots,3)$, the symmetry properties in Eq. (16) (with $\Xi=\Upsilon=0$) lead to the following dissipation function

$$\Psi = \int d\mathbf{r} \left[\frac{\zeta_u}{2} (\dot{u}_x^2 + \dot{u}_y^2) + \frac{\zeta_s}{2} \dot{s}^2 + \frac{\mu}{2} (\tau_0 \dot{s} - \dot{\tau}_1)^2 + \frac{\nu}{2} (\tau_3 \dot{s} - \dot{\tau}_2)^2 + \frac{\nu}{2} (\tau_2 \dot{s} + \dot{\tau}_3)^2 \right]. \tag{17}$$

The first two terms are similar to those in Eq. (3). In the above, we have introduced the positive friction coefficients μ and ν that are related to the microscopic structure, such as the lattice topology of the odd microdevice.²⁶⁾ The terms that can be obtained by replacing $\dot{s} \rightarrow -\dot{s}$ are not included because one of the rotational directions of \dot{s} is specified as in the first model. We also do not consider the self-driven terms $(\tau_{\alpha}\dot{s} - \dot{\tau}_{\alpha})^2$ since they do not contribute to the odd elasticity. By adding Eqs. (15) and (17), the Rayleighian of the continuum model is now given by $\mathcal{R} = \dot{F} + \Psi$.

Next, we functionally minimize the Rayleighian \mathcal{R} with respect to \dot{u}_x , \dot{u}_y , and \dot{s} , and obtain the following set of coupled equations

$$\zeta_{u}\dot{u}_{x} - \partial_{i}\sigma_{xi} - \mu\partial_{y}(\tau_{0}\dot{s} - \dot{\tau}_{1})$$
$$-\nu[\partial_{x}(\tau_{3}\dot{s} - \dot{\tau}_{2}) + \partial_{y}(\tau_{2}\dot{s} + \dot{\tau}_{3})] = 0, \tag{18}$$

$$\zeta_{\mu}\dot{u}_{\nu} - \partial_{i}\sigma_{\nu i} + \mu\partial_{x}(\tau_{0}\dot{s} - \dot{\tau}_{1})$$

$$+\nu[\partial_{\nu}(\tau_{3}\dot{s} - \dot{\tau}_{2}) - \partial_{\nu}(\tau_{2}\dot{s} + \dot{\tau}_{3})] = 0, \tag{19}$$

$$\zeta_s \dot{s} + \mu(\tau_0^2 \dot{s} - \tau_0 \dot{\tau}_1) + \nu[(\tau_2^2 + \tau_3^2) \dot{s} - \dot{\tau}_2 \tau_3 + \tau_2 \dot{\tau}_3] = f. \tag{20}$$

The above equations are nonlinear and cannot be solved analytically. To simplify the situation, we focus on the small-strain limit for which the equations can be linearized. Then Eq. (20) becomes $\zeta_s \dot{s} \approx f$. By substituting this relation into Eqs. (18) and (19), we obtain the matrix equation of Eq. (7) in which the friction matrix operator Γ is

$$\Gamma = \begin{pmatrix} \zeta_u - \mu \partial_y^2 - \nu \nabla^2 & 0 \\ 0 & \zeta_u - \mu \partial_x^2 - \nu \nabla^2 \end{pmatrix}, \tag{21}$$

where ∇^2 is the 2D Laplacian, and the components of the generalized elastic matrix operator **E** are

$$E_{11} = (K+G)\partial_x^2 + G\partial_y^2 + (f\mu/\zeta_s)\partial_x\partial_y, \tag{22}$$

$$E_{12} = K \partial_x \partial_y + (f \mu / \zeta_s) \partial_y^2 + (f \nu / \zeta_s) \nabla^2, \tag{23}$$

$$E_{21} = K \partial_x \partial_v - (f \mu / \zeta_s) \partial_v^2 - (f \nu / \zeta_s) \nabla^2, \tag{24}$$

$$E_{22} = (K+G)\partial_{\nu}^2 + G\partial_{x}^2 - (f\mu/\zeta_s)\partial_{x}\partial_{y}.$$
 (25)

We now apply the Fourier transform of the obtained linearized equation. For an isotropic 2D material, one can choose the *x*-direction as the direction of perturbation without loss of generality. By using the solution of the form $u_x \sim e^{i(qx-\omega t)}$ and $u_y \sim e^{-i\omega t}$, where q is wavenumber and ω is

frequency, Eq. (7) can be rewritten in terms of the Fourier components as

$$i\omega \begin{pmatrix} \zeta_u + \nu q^2 & 0 \\ 0 & \zeta_u + (\mu + \nu)q^2 \end{pmatrix} \begin{pmatrix} u_x \\ u_y \end{pmatrix}$$

$$= q^2 \begin{pmatrix} K + G & f\nu/\zeta_s \\ -f(\mu + \nu)/\zeta_s & G \end{pmatrix} \begin{pmatrix} u_x \\ u_y \end{pmatrix}. \tag{26}$$

The two odd elastic moduli can now be identified as $A = -f\mu/\zeta_s$ and $K^o = f\nu/\zeta_s$ (the minus sign of A is corrected as in Ref. 16), which are both proportional to the nonequilibrium force f and the ratio between the friction coefficients (either μ/ζ_s or ν/ζ_s). Notice that A and K^o can take both positive and negative signs depending on the sign of f. Hence we have provided a clear physical meaning of the odd elasticity in a nonequilibrium system within the OVP framework. This is the main outcome of our formulation. Note that the q^2 -dependence in the friction matrix of Eq. (26) is a new contribution.

In the long-wavelength limit where the q-dependence on the left-hand side of Eq. (26) can be neglected, we obtain the following approximate dispersion relation

$$i\omega \approx \frac{q^2}{\zeta_u} \left[\frac{K}{2} + G \pm \sqrt{\left(\frac{K}{2}\right)^2 - \frac{f^2 \mu \nu}{\zeta_s^2} - \left(\frac{f \nu}{\zeta_s}\right)^2} \right].$$
 (27)

The instability occurs when the real part of $i\omega$ becomes negative, i.e., when

$$\frac{f\mu}{\zeta_s} < -\frac{KG + G^2 + (f\nu/\zeta_s)^2}{f\nu/\zeta_s}.$$
 (28)

Moreover, an exceptional point exists when the two eigenvalues of Eq. (27) coincide. The oscillatory dynamics takes place when $f\mu/\zeta_s > (K/2)^2/(f\nu/\zeta_s) - f\nu/\zeta_s$. ¹⁵⁾

To summarize, we have obtained dynamical equations for a nonequilibrium active system with odd elasticity within the framework of OVP. The nonreciprocal equations result from the elimination of the extra variable that is conjugate to the nonequilibrium driving force. We have explicitly shown that the odd elastic moduli are proportional to the nonequilibrium force and the friction coefficients.

The concept of the extra nonequilibrium variable and the resulting nonreciprocality can also be seen in the studies for micropolar fluid²⁵⁾ and odd viscosity.^{27,28)} In these works, the extra coordinate represents either the rotating field or the intrinsic rotor. Different from the approach in Ref. 13, we followed the framework of OVP to obtain the active dynamics by using the extra nonequilibrium variable. This clarifies the energy injection process from the active sources and provides the explicit expressions of the odd elastic moduli.

For general nonreciprocal systems, f and s in the free energy function do not need to stand for specific rotational quantities in the configuration space. Any set of generalized forces with their conjugated variables can be chosen as long as they drive the system away from the equilibrium and give rise to a rotational trajectory in the state space. In an enzymatic system, for example, f and s correspond to the

chemical potential difference and the chemical reaction variable, respectively. $^{19,21,29)}$ In the future, the microscopic origin of the active quantities such as f, μ , and ν in our continuum model needs to be further clarified by using various coarse-graining methods.

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