Statistical formulation of Onsager-Machlup variational principle

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Onsager's variational principle (OVP) provides us with a systematic way to derive dynamical equations for various soft matter and active matter. By reformulating the Onsager-Machlup variational principle (OMVP), which is a time-global principle, we propose a new method to incorporate thermal fluctuations. To demonstrate the utility of the statistical formulation of OMVP (SOMVP), we obtain the diffusion constant of a Brownian particle embedded in a viscous fluid only by maximizing the modified Onsager-Machlup integral for the surrounding fluid. We also apply our formulation to a Brownian particle in a steady shear flow, which is a typical example of a non-equilibrium system. Possible extensions of our formulation to internally driven active systems are discussed.

Introduction.– Onsager's variational principle (OVP) is useful to obtain the governing equations of irreversible dynamics in soft matter such as polymers, colloidal suspensions, and liquid crystals [1–4]. In this principle, we construct a functional quantity called Rayleighian by summing up the dissipation function and the change rate of free energy. Minimization of Rayleighian under appropriate constraints provides us with overdamped deterministic equations that describe the change rate of the state variables. The obtained equations automatically satisfy Onsager's reciprocal relations and the second law of thermodynamics. To further include thermal fluctuations, noise terms are added to the deterministic equations, and their statistical properties should be determined by the fluctuation-dissipation theorem [4].

For time-evolving processes under thermal fluctuations, one can use the Onsager-Machlup (OM) integral to discuss the path probability [5, 6]. The OM integral has been used in various problems, such as structural transitions of protein folding [7], chemical kinetic models [8], and active matter systems [9–12]. Moreover, a path integral representation of fluctuating hydrodynamics can also be provided by the OM integral [13–16].

Using OM integral, Doi *et al.* proposed Onsager-Machlup variational principle (OMVP) [17, 18]. In contrast to OVP, which is a time-local principle, OMVP is a time-global variational principle, and it allows us to obtain the most probable trajectory over a long time. In particular, OMVP is useful for finding accurate solutions to the time evolution equations when the solutions involve unavoidable errors due to the precision limit of numerical calculation and can determine long-time behaviors such as steady states. However, the statistical property of the trajectories and the effects of thermal fluctuations, such as the mean square displacement of a Brownian particle, cannot be directly obtained within OMVP, which is left as an important issue especially for active systems [17, 18].

In this Letter, we propose a statistical formulation of OMVP (SOMVP) by taking into account thermal fluctuations of stochastic systems. We introduce an *observable* that is determined by the system variables and allows the OM integral to explore trajectories deviating far from the most probable path. We propose a modified OM integral that should be maximized to obtain the cumulant-generating function (CGF) of the observable. To demonstrate the usefulness of SOMVP, we calculate the diffusion constant of a Brownian particle embedded in a viscous fluid only by optimizing the modified OM integral. Notably, the obtained diffusion constant recovers the Stokes-Einstein relation or the fluctuation-dissipation relation without any additional requirements. To further show that SOMVP can be applied to non-equilibrium phenomena, we calculate the CGF of a Brownian particle subjected to a steady shear flow. We shall also argue several possibilities for applying SOMVP for internally driven active matter systems.

OVP and OMVP.– Let us consider a system that is described by the state variable $\mathbf{x}(\mathbf{r})$ and its change rate $\mathbf{v}(\mathbf{r})$. Here, both $\mathbf{x}(\mathbf{r})$ and $\mathbf{v}(\mathbf{r})$ are functions of space $\mathbf{r}$ and they are independent of each other. According to OVP, we minimize Rayleighian $R[\mathbf{v}(\mathbf{r}), \mathbf{x}(\mathbf{r})]$ with respect to $\mathbf{v}(\mathbf{r})$ to obtain the deterministic equations for $\mathbf{x}(\mathbf{r})$ and $\mathbf{v}(\mathbf{r})$ [3, 4]. The advantage of OVP is that the obtained equations automatically satisfy Onsager's reciprocal relations and the second law of thermodynamics. The Rayleighian is given by $R = \Phi + \dot{F} + C$, where Φ is the dissipation function, $\dot{F}$ is the rate of change of free energy, and C represents various constraints that are included by using Lagrange multipliers. Notice that OVP determines the instantaneous change rate $\mathbf{v}(\mathbf{r})$ at state $\mathbf{x}(\mathbf{r})$.

Next, we consider the time dependence of the considered variables, $\mathbf{x}(\mathbf{r}, t)$ and $\mathbf{v}(\mathbf{r}, t)$. We introduce the OM integral that is given by the time integral of the

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Rayleighian [5, 6]

$$O[\mathbf{v}(\mathbf{r},t),\mathbf{x}(\mathbf{r},t)] \\ = \int_0^\tau dt\,(R[\mathbf{v}(\mathbf{r},t),\mathbf{x}(\mathbf{r},t)] - R_{\min}[\mathbf{x}(\mathbf{r},t)]), \quad (1)$$

where $R_{\min}$ is the minimum value of R in $\mathbf{v}$-space, i.e., $R_{\min}[\mathbf{x}(\mathbf{r})] = \min_{\mathbf{v}(\mathbf{r})} R[\mathbf{v}(\mathbf{r}),\mathbf{x}(\mathbf{r})]$, and τ is the duration time. As a global variational principle, OMVP states that nature chooses the path that minimizes the OM integral [17].

Statistical formulation of OMVP.– In the presence of thermal fluctuations, stochastic dynamics of $\mathbf{x}(\mathbf{r},t)$ and $\mathbf{v}(\mathbf{r},t)$ involve uncertainty. To discuss the probability of a trajectory, Onsager and Machlup considered the path probability under the initial condition $\mathbf{x}(\mathbf{r},0) = \mathbf{x}_0(\mathbf{r})$ such that [5, 6]

$$P[\mathbf{v}(\mathbf{r},t),\mathbf{x}(\mathbf{r},t);\mathbf{x}_0(\mathbf{r})] \sim \exp\left(-\frac{O[\mathbf{v}(\mathbf{r},t),\mathbf{x}(\mathbf{r},t)]}{2k_{\mathrm{B}}T}\right), \quad (2)$$

where k_{B} is the Boltzmann constant and T is the temperature of the system.

We now propose a statistical formulation of OMVP (SOMVP), where we consider a stochastic *observable* X that is determined by $\mathbf{v}(\mathbf{r},t)$ and $\mathbf{x}(\mathbf{r},t)$. Examples of X are the trajectory of $\mathbf{x}(\mathbf{r},t)$, the time average of $\mathbf{v}(\mathbf{r},t)$ or $\mathbf{x}(\mathbf{r},t)$ (see later examples), the irreversibility [19], and the edge current [20]. Let us consider the cumulant generating function (CGF) of the observable X introduced by

$$K_X(q) = \ln\langle\exp(qX)\rangle. \quad (3)$$

In the above, the statistical average of a stochastic quantity A is calculated by $\langle A\rangle = \int \mathcal{D}\mathbf{v}\mathcal{D}\mathbf{x}\,AP$, where P is the path probability in Eq. (2) and $\int \mathcal{D}\mathbf{v}\mathcal{D}\mathbf{x}$ indicates the path integral over all the trajectories $\mathbf{v}(\mathbf{r},t)$ and $\mathbf{x}(\mathbf{r},t)$. Then, the n-th cumulant can be obtained from the CGF as

$$\langle X^n\rangle_{\mathrm{c}} = \left.\frac{d^n}{dq^n}K_X(q)\right|_{q=0}. \quad (4)$$

By substituting Eq. (2) into Eq. (3) and employing the saddle-point approximation used in the large deviation theory [21, 22], the approximate CGF is given by

$$K_X(q) \approx N + \max_{\mathbf{v}(\mathbf{r},t),\mathbf{x}(\mathbf{r},t);\mathbf{x}_0(\mathbf{r})} \Omega[\mathbf{v}(\mathbf{r},t),\mathbf{x}(\mathbf{r},t)], \quad (5)$$

$$\Omega[\mathbf{v}(\mathbf{r},t),\mathbf{x}(\mathbf{r},t)] = qX - \frac{O[\mathbf{v}(\mathbf{r},t),\mathbf{x}(\mathbf{r},t)]}{2k_{\mathrm{B}}T} + \Gamma, \quad (6)$$

where N is the normalization factor determined by the condition $K_X(0) = 0$. Equations (5) and (6) are the proposed SOMVP. In the above, Ω is the modified OM integral to be maximized with respect to both $\mathbf{v}(\mathbf{r},t)$ and $\mathbf{x}(\mathbf{r},t)$ under the given initial condition $\mathbf{x}_0(\mathbf{r})$. As shown in Eq. (6), the original OM integral O is modified by the observable X, which allows the trajectories to deviate from the most probable path. Such a path exploration is essential to calculate the CGF in the current framework. In Eq. (6), a trivial connection between $\mathbf{v}$ and $\mathbf{x}$ is introduced by the term Γ since the maximization in Eq. (5) is taken with respect to both $\mathbf{v}$ and $\mathbf{x}$. More specifically, we use $\Gamma = \int_0^\tau dt\,H_i(\dot{x}_i - v_i)$, where $\mathbf{H}$ is the Lagrange multiplier and we have used the Einstein summation convention.

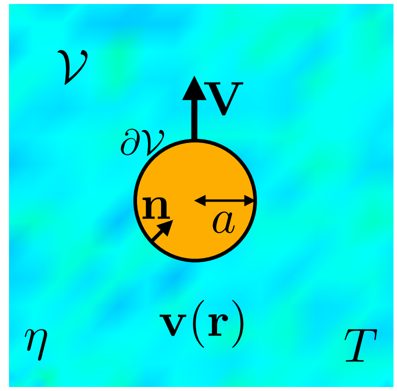

FIG. 1. A spherical particle of radius a is embedded in a three-dimensional viscous fluid with viscosity η and temperature T. The fluid velocity field $\mathbf{v}(\mathbf{r})$ fluctuates due to thermal fluctuations. The velocity of the particle $\mathbf{V}$ is subjected to the random forces of the surrounding fluid. The space filled with the fluid is denoted by $\mathcal{V}$, whereas the surface of the particle is denoted by $\partial\mathcal{V}$.

Brownian particle in a viscous fluid.– As the simplest demonstration of SOMVP, we consider a Brownian particle embedded in a viscous fluid in the presence of thermal fluctuations. In the previous works, Langevin equation of a Brownian particle was derived by solving the boundary problem of the fluctuating hydrodynamics [14, 15]. Later, the drag coefficient of a Brownian particle was calculated by connecting the fluctuating hydrodynamics and Hamiltonian dynamics [16]. Although the fundamental concept of integrating the fluid degrees of freedom and casting it to the motion of a Brownian particle is similar to the previous approaches, we show below a more systematic derivation of the particle diffusion constant by using SOMVP.

As shown in Fig. 1, we consider a spherical particle of radius a immersed in a fluid with viscosity η and temperature T. First, the dissipation function of the fluid moving with the velocity $\mathbf{v}(\mathbf{r})$ is given by [3, 4]

$$\Phi = \frac{\eta}{4}\int_{\mathcal{V}} d^3r\,(\partial_i v_j + \partial_j v_i)^2, \quad (7)$$

where the integral is over the three-dimensional fluid region $\mathbf{r} = (x,y,z) \in \mathcal{V}$ and $i,j = x,y,z$. Second, there is no free energy (potential energy) in the current problem.

Third, we employ two constraints on the fluid properties: the incompressibility condition and the stick boundary condition at the surface of the particle [23]. These conditions can be included by the following terms

$$C_1 = -\int_{\mathcal{V}} d^3r\, p\partial_i v_i, \tag{8}$$

$$C_2 = \int_{\partial\mathcal{V}} dS\, g_i(v_i - V_i), \tag{9}$$

where p and $\mathbf{g}$ are the Lagrange multipliers, $\mathbf{V}$ is the particle velocity, and $\partial\mathcal{V}$ represents the surface of the particle. Notice that the particle displacement is given by $X_i(\tau) = \int_0^\tau dt\, V_i(t)$.

The Rayleighian is simply given by $R = \Phi + C_1 + C_2$, and the OM integral is obtained by using Eq. (1). In the current problem, the term $R_{\min}$ can be neglected since it does not depend on $\mathbf{x}$. We consider the CGF $K_{\mathbf{X}}(\mathbf{q})$ of the particle displacement $\mathbf{X}(\tau)$ up to time τ, i.e., $K_{\mathbf{X}}(\mathbf{q}) = \ln\langle \exp[q_i X_i(\tau)]\rangle$. Since the OM integral does not depend on $\mathbf{X}(t)$, we obtain the modified OM integral Ω by setting $\Gamma = 0$ in Eq. (6).

Following Eq. (5) and maximizing Ω with respect to $\mathbf{v}$, $\mathbf{V}$, p, and $\mathbf{g}$, i.e., $\delta\Omega = 0$, we obtain the following governing equations:

$$\eta\nabla^2 v_i - \partial_i p = 0, \quad \partial_i v_i = 0 \quad (\mathbf{r} \in \mathcal{V}), \tag{10}$$

$$v_i = V_i \quad (\mathbf{r} \in \partial\mathcal{V}), \tag{11}$$

$$q_i - \frac{1}{2k_{\mathrm{B}}T}\int_{\partial\mathcal{V}} dS\, n_j\sigma_{ij} = 0. \tag{12}$$

In Eq. (12), the stress tensor is given by $\sigma_{ij} = \eta(\partial_i v_j + \partial_j v_i) - p\delta_{ij}$, where p is the pressure. As shown in Fig. 1, $\mathbf{n}$ is the unit normal vector pointing from the fluid to the particle, and the fluid velocity is assumed to vanish at infinity, $\mathbf{v}(r \to \infty) = 0$. In the above equations, we first need to solve the Stokes equations to obtain the Stokes' law. Then, we use Eq. (12) to calculate $\mathbf{V}$.

Let us briefly summarize the solutions to Stokes equation in the presence of a spherical particle [24, 25]. One solution is the Stokeslet $v_i^{\mathrm{S}} = f_j G_{ij}/(8\pi\eta)$ and $p^{\mathrm{S}} = f_i P_i/(8\pi)$, where f_i are coefficients fixed by the boundary condition, $G_{ij} = r^{-1}\delta_{ij} + r^{-3}r_i r_j$ and $P_i = 2r^{-3}r_i$ with $r = |\mathbf{r}|$. The other is the potential dipole solution $v_i^{\mathrm{PD}} = h_j\nabla^2 G_{ij}/(8\pi\eta)$ with unknown coefficients h_i and $p^{\mathrm{PD}} = p_0$, where p_0 is a uniform pressure and is assumed to be zero. Adding these two solutions, $v_i = v_i^{\mathrm{S}} + v_i^{\mathrm{PD}}$ and $p = p^{\mathrm{S}} + p^{\mathrm{PD}}$, we obtain the solution that satisfies the boundary condition at the particle surface. After some calculation, we obtain $f_i = 6\pi\eta a V_i$ and $h_i = a^2 f_i/6$ by using Eq. (11). Then, the surface integral of the stress tensor becomes $\int_{\partial\mathcal{V}} dS\, n_j\sigma_{ij} = f_i$ [24, 25]. By using Eq. (12), the particle velocity $\mathbf{V}$ can be obtained as

$$V_i = \frac{k_{\mathrm{B}}Tq_i}{3\pi\eta a}. \tag{13}$$

Next, we calculate the maximized Ω by substituting the above results. The dissipation function in Eq. (7) now becomes $\Phi = \int_{\partial\mathcal{V}} dS\, n_i\sigma_{ij}v_j/2 = V_i f_i/2$ [24], whereas the constraint terms obviously disappear; $C_1 = C_2 = 0$.

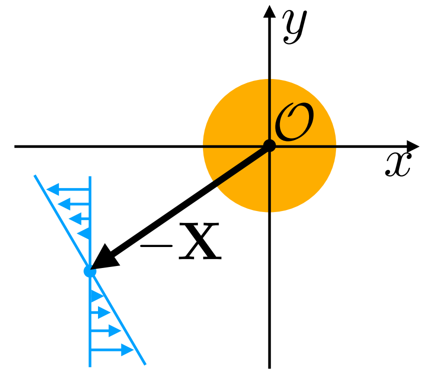

FIG. 2. A Brownian particle in a steady shear flow. We use the coordinate system in which the particle is fixed such that the origin of the space, $\mathcal{O}$, is fixed at the center of the particle. On the other hand, the center of the shear flow (blue circle) is located at $-\mathbf{X}$, where $\mathbf{X}$ is the particle displacement.

Substituting these results into Ω in Eq. (6), we obtain the CGF as

$$K_{\mathbf{X}}(\mathbf{q}) = q_i^2 D\tau, \tag{14}$$

where $D = k_{\mathrm{B}}T/(6\pi\eta a)$ is the diffusion coefficient that recovers the Stokes-Einstein relation without additional requirements for fluctuations. (Notice that N in Eq. (5) vanishes because $K_{\mathbf{X}}(0) = 0$.) Hence, the second-order cumulant or the mean squared displacement of the particle becomes

$$\langle X_i X_j\rangle_{\mathrm{c}} = 2D\tau\delta_{ij}, \tag{15}$$

recovering the required statistical property of the Brownian motion [4].

The above example demonstrates that we can use SOMVP to integrate out the stochastic dynamics of the surrounding fluid and discuss the statistical properties of the macroscopic observable. To further show that SOMVP also works in non-equilibrium systems, we calculate the CGF of a Brownian particle subjected to a steady shear flow.

Brownian particle in a shear flow.– Next, we consider a Brownian particle in a steady shear flow, as shown in Fig. 2. We employ a coordinate system in which the particle is fixed and the center of the shear flow is located at $-\mathbf{X}$, where $\mathbf{X}$ is the particle displacement from the center of the shear flow. In this problem, the flow field can be decomposed into the shear part and the remainig part as $\mathbf{v}^{\mathrm{shear}} + \mathbf{v}$, where $v_i^{\mathrm{shear}} = \dot{\gamma}_{ij}(r_j + X_j)$ and $\dot{\gamma}_{ij}$ is the shear rate tensor satisfying $\dot{\gamma}_{ii} = 0$.

We consider the Rayleighian for the remaining part $\mathbf{v}$. The dissipation function is the same as in Eq. (7), and the incompressibility condition is also given by Eq. (8). The boundary condition $\mathbf{v}^{\mathrm{shear}} + \mathbf{v} = \mathbf{V}$ on the particle surface is taken into account by the following term

$$C_2' = \int_{\partial\mathcal{V}} dS\, g_i[v_i - V_i + \dot{\gamma}_{ij}(r_j + X_j)]. \tag{16}$$

Ignoring $R_{\min}$ in Eq. (1) as before, we obtain the modified OM integral Ω in Eq. (6) by using the Rayleighian $R = \Phi + C_1 + C_2'$. Unlike the previous case, Ω is a functional of both $\mathbf{v}$ and $\mathbf{V}$ and the particle position $\mathbf{X}$. Due to this dependence on $\mathbf{X}$, we need an additional term $\Gamma = \int_0^\tau dt H_i(\dot{X}_i - V_i)$ in Ω, ensuring $\dot{\mathbf{X}} = \mathbf{V}$. Maximizing Ω with respect to $\mathbf{v}$, $\mathbf{V}$, $\mathbf{X}$, p, $\mathbf{g}$, and $\mathbf{H}$, i.e., $\delta\Omega = 0$, we obtain Eq. (10) and the following set of equations:

$$v_i = V_i - \dot{\gamma}_{ij}(r_j + X_j) \quad (\mathbf{r} \in \partial\mathcal{V}), \tag{17}$$

$$q_i - H_i - \frac{1}{2k_{\mathrm{B}}T}\int_{\partial\mathcal{V}} dS\, n_j\sigma_{ij} = 0, \tag{18}$$

$$-\dot{H}_i + \dot{\gamma}_{ji}(q_j - H_j) = 0, \quad H_i(\tau) = 0. \tag{19}$$

In the current problem, the solution to the Stokes equation in Eq. (10) can be constructed as $v_i = v_i^{\mathrm{S}} + v_i^{\mathrm{PD}} + v_i^{\mathrm{SD}} + v_i^{\mathrm{PQ}}$, where v_i^{S} and v_i^{PD} are the flows due to the Stokeslet and the potential dipole solution, respectively, as before. The additional terms are the Stokes dipole solution $v_i^{\mathrm{SD}} = \xi_{jk}\partial_k G_{ij}$, $p^{\mathrm{SD}} = \eta\xi_{ij}\partial_j P_i$ and the potential quadrupole solution $v_i^{\mathrm{PQ}} = \zeta_{jk}\partial_k\nabla^2 G_{ij}$, $p^{\mathrm{PQ}} = p_0$, where the coefficients ξ_{ij} and ζ_{ij} should be fixed by the boundary condition in Eq. (17) [24, 25]. After some calculation, all the coefficients can be determined as

$$f_i = 6\pi\eta a(V_i - \dot{\gamma}_{ij}X_j), \quad h_i = a^2 f_i/6, \tag{20}$$

$$\xi_{ij} = 5\dot{\gamma}_{ij}^{\mathrm{S}}/3 + \dot{\gamma}_{ij}^{\mathrm{A}}, \quad \zeta_{ij} = 18a^2\xi_{ij}/5, \tag{21}$$

where $\dot{\gamma}_{ij}^{\mathrm{S}} = (\dot{\gamma}_{ij} + \dot{\gamma}_{ji})/2$ and $\dot{\gamma}_{ij}^{\mathrm{A}} = (\dot{\gamma}_{ij} - \dot{\gamma}_{ji})/2$ are the symmetric and antisymmetric parts of the shear rate tensor, respectively.

By evaluating the surface integral of the stress tensor, Eq. (18) becomes the following differential equation:

$$V_i = \dot{\gamma}_{ij}X_j + 2D(q_i - H_i), \tag{22}$$

where $V_i = \dot{X}_i$. Hereafter, we consider the simplest shear flow in the xy-plane, $\dot{\gamma}_{ij} = \dot{\gamma}\delta_{ix}\delta_{jy}$. To find the optimum trajectory $\mathbf{X}(t)$, we first solve Eq. (19) to obtain $H_x = H_z = 0$ and $H_y = \dot{\gamma}q_x(t-\tau)$, where we have used the final condition $H_i(\tau) = 0$. With these solutions, Eq. (22) can be rewritten as $V_x = \dot{\gamma}Y + 2Dq_x$, $V_y = -2D(H_y - q_y)$, and $V_z = 2Dq_z$. Solving these equations with the initial condition $\mathbf{X}_0 = 0$, we get the optimum trajectory as

$$X(t) = 2D[q_x t - \dot{\gamma}^2 q_x(t^3/6 - t^2\tau/2) + \dot{\gamma}q_y t^2/2], \tag{23}$$

$$Y(t) = 2D[q_y t - \dot{\gamma}q_x(t^2/2 - t\tau)], \tag{24}$$

$$Z(t) = 2Dq_z t. \tag{25}$$

At this stage, the dissipation function becomes $\Phi = (V_i - \dot{\gamma}_{ij}X_j)f_i/2 + \Phi_0$, where $\Phi_0 = -\dot{\gamma}_{jk}\int_{\partial\mathcal{V}} dS\, n_i\sigma_{ij}r_k/2$ does not depend on $\mathbf{q}$. Substituting the optimal trajectories in Eqs. (23)-(25), we obtain

$$\Phi - \Phi_0 = 2k_{\mathrm{B}}TD[q_x^2 + q_z^2 + \{q_y - \dot{\gamma}q_x(t-\tau)\}^2]. \tag{26}$$

Then, the CGF in the presence of a shear flow becomes

$$K_{\mathbf{X}}(\mathbf{q}) = D[q_i^2\tau + q_x q_y\dot{\gamma}\tau^2 + q_x^2\dot{\gamma}^2\tau^3/3], \tag{27}$$

where we have fixed N to satisfy $K_X(0) = 0$ and Φ_0 has been cancelled by N.

Finally, the second-order cumulants are obtained from the CGF as

$$\langle X^2\rangle_{\mathrm{c}} = 2D\tau + 2D\dot{\gamma}^2\tau^3/3, \tag{28}$$

$$\langle Y^2\rangle_{\mathrm{c}} = \langle Z^2\rangle_{\mathrm{c}} = 2D\tau, \tag{29}$$

$$\langle XY\rangle_{\mathrm{c}} = D\dot{\gamma}\tau^2. \tag{30}$$

These results recover those in Refs. [26–29] and Eq. (28) is different from Eq. (15) due to the shear flow. Notice that the correction term is proportional to τ^3 [27, 28]. As shown in Eq. (30), the shear flow also leads to the cross-correlation between X and Y, being proportional to τ^2. From Eq. (27), we further notice that the higher order cumulants vanish, i.e., $\langle\mathbf{X}^n\rangle_{\mathrm{c}} = 0$ for $n \geq 3$. The above example demonstrates that SOMVP can be applied not only for thermal equilibrium systems but also for out-of-equilibrium systems driven by external flows.

Summary and Discussion.– In this Letter, we have proposed a statistical formulation of OMVP (SOMVP) as shown in Eqs. (5) and (6). Using this method, one can systematically obtain the cumulants of any stochastic observable. We showed that SOMVP reproduces the established results for Brownian particles in thermal equilibrium [see Eqs. (15)] and in out-of-equilibrium cases such as with an applied shear flow [see Eqs. (27)]. SOMVP can be used to other stochastic systems that cannot be described by trivial governing equations due to the complicated geometry of the problem.

The other advantage of SOMVP is that we can reuse the Rayleighian considered for different soft matter systems [4], such as polymers [18], liquid crystals [30], membranes surrounded by a bulk fluid [31, 32]. Moreover, various boundary conditions can be systematically taken into account in SOMVP by using Lagrange multipliers. On the other hand, one limitation of the current SOMVP is that it cannot deal with active non-thermal fluctuations. To include non-equilibrium fluctuations, a further generalization of SOMVP is required.

In this work, we have applied our new formulation to an externally driven non-equilibrium system under a shear flow. The method of SOMVP can also be used for internally driven active systems in which additional work W is generated by an active element owing to energy input or chemical reactions. In this case, the Rayleighian is extended to $R = \Phi + \dot{F} + \dot{W} + C$, which includes the power of active work $\dot{W}$ [10]. Following a similar procedure, one can systematically discuss the statistical properties of active systems, such as the diffusion constant of internally driven objects.

For example, we can consider a chemically activated Janus particle for which chemical reactions take place on half of the particle surface [33, 34]. We use the volume fraction of the chemical product $\phi(\mathbf{r})$ to express the dissipation function and the free energy of a binary mixture. To describe the chemical reaction occurring on half of the particle surface $\partial_+\mathcal{V}$, we use the fixed-concentration condition, $\phi(\mathbf{r}) = 1$ ($\mathbf{r} \in \partial_+\mathcal{V}$). Such a formulation would help to understand the enhanced self-diffusion of a catalytic enzyme molecule [35].

Another example is the observed active diffusion in living cells [36] driven by active force dipoles [37, 38]. The power due to active force dipoles can be described by $\dot{W} = cm \int_{\mathcal{V}} d^3r\, e_i(\mathbf{r}) e_j(\mathbf{r}) \partial_i v_j(\mathbf{r})$, where c is the concentration of the dipole, m is the magnitude of the force dipole, and $\mathbf{e}$ is a unit vector representing the direction of a dipole. Adding $\dot{W}$ and the dissipation function describing rotational motions of dipoles to the Rayleighian, we can discuss the Brownian motion of a tracer particle in active fluids. The details will be our future work.

In this work, we have obtained the CFG by analytically solving the obtained dynamical equations derived from SOMVP. When such an approach is difficult, one can also numerically optimize the modified OM integral. In such a situation, it may be useful to use techniques in machine learning, such as reinforcement learning [39, 40].

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