Microscopic Cutoff Dependence of an Entropic Force in Interface Propagation of Stochastic Order Parameter Dynamics

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The steady propagation of a $(d-1)$-dimensional planer interface in d-dimensional space is studied by analyzing mesoscopic nonconserved order parameter dynamics with two local minima under the influence of thermal noise. In this analysis, an entropic force generating interface propagation is formulated using a perturbation method. It is found that the entropic force singularly depends on an ultraviolet cutoff when $d \geq 2$. The theoretical calculation is confirmed by numerical simulations with $d = 2$. The result means that an experimental measurement of the entropic force provides an estimation of the microscopic cutoff of the mesoscopic description.

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Introduction.—Macroscopic dynamics in nature are often described by deterministic equations. If a class of phenomena is found to be described by a simple deterministic equation with a few parameters, the phenomena can be studied by analyzing the universal equation. Fluid dynamics is a typical example of the success of such an approach [1]. However, there are cases in which simple deterministic equations cannot be found. For example, the macroscopic motion of locally conserved quantities in one or two dimensions cannot be described by a hydrodynamic equation with finite transportation coefficients [2]. Another example is the dynamical behavior near a critical point, where transport coefficients show a singular behavior [3,4].

Even in such cases, a mesoscopic model with thermal noise can describe the macroscopic dynamical behavior. For example, fluctuating hydrodynamics can correctly describe the singular behavior of hydrodynamics in low dimensions [2]. The dynamical behavior near a critical point also can be described by the Ginzburg-Landau model with thermal noise [5]. Here, fluctuations modify the mean-field properties given by the mesoscopic free energy, which includes the transition type [6] as well as the critical exponents [5,7]. These examples show that fluctuation effects at mesoscopic scales lead to the renormalization of model parameters and that the lack of a simple macroscopic equation is connected to the infrared divergence of renormalized parameters. In such a case, phenomena observed at macroscopic scales cannot be separated from those at mesoscopic scales.

The purpose of this Letter is to report another violation of this scale separation, which is qualitatively different from the previously known cases. We study a steady propagation of a $(d-1)$-dimensional planer interface separating two phases in d dimensions by analyzing the mesoscopic nonconserved order parameter dynamics with weak thermal noise. One remarkable finding is that the propagation velocity depends on the noise intensity, as already reported in the case $d = 1$ [8]. This means that the noise modifies the average dynamic behavior. Although this is an interesting phenomenon, the mechanism behind it is simple. The driving force of the interface motion is the free energy difference between the two phases, which contains the entropic contribution in addition to the mesoscopic free energy given by the mesoscopic model. Here, the entropic contribution is expressed by fluctuation properties coming from the noise. Thus, if there is no special symmetry between the two phases, the propagation velocity depends on the noise intensity. In a special case where the mesoscopic free energy takes the same value in the two phases, the propagation occurs only as a result of the entropic force. The behavior can be understood by the renormalization of the mesoscopic free energy. The main message of this Letter is that the entropic force diverges when an ultraviolet cutoff goes to infinity for $d \geq 2$.

This result means that the propagation velocity driven by the entropic force depends on the microscopic cutoff. In other words, the mesoscopic description cannot be separated from a more microscopic system. We demonstrate this result using a theoretical calculation of the entropic force. Furthermore, we confirm this claim by performing numerical simulations of the order parameter dynamics with noise. We expect that this singular behavior is also observed in experiments such as interface motion in a spin-crossover complex [9]. Surprisingly, an experimental measurement of the entropic force provides an estimation of the microscopic cutoff of the mesoscopic description.

Setup.—For simplicity, we present the system in two dimensions. The generalization to the other dimensions is straightforward. Let $\boldsymbol{r} = (x, y) \in \mathbb{R}^2$ be a position in a two-dimensional region $D \equiv [-L, L] \times [0, L_y]$. L and L_y are sufficiently large and L is assumed to be infinity in theoretical arguments. We define a real scalar order parameter field $\phi(\boldsymbol{r}, t)$ in the region D. The free energy functional of ϕ is given by

$$\mathcal{F}(\phi) = \int_D d^2\boldsymbol{r}\left\{f(\phi) + \frac{\kappa}{2}[(\partial_x\phi)^2 + (\partial_y\phi)^2]\right\}, \quad (1)$$

where $f(\phi)$ is a mesoscopic free energy density and κ is a constant characterizing the interface energy. Following the Onsager principle, we assume that the dynamics of $\phi(\boldsymbol{r}, t)$ is described by

$$\partial_t\phi = -\Gamma\frac{\delta\mathcal{F}}{\delta\phi} + \eta, \quad (2)$$

where Γ is a constant representing the mobility and η is Gaussian white noise satisfying $\langle\eta(\boldsymbol{r}, t)\rangle = 0$ and the fluctuation-dissipation relation

$$\langle\eta(\boldsymbol{r}, t)\eta(\boldsymbol{r}', t')\rangle = 2\Gamma T\delta(\boldsymbol{r} - \boldsymbol{r}')\delta(t - t'). \quad (3)$$

This model has been referred to as "Model A" [5], which describes the nonconserved order parameter dynamics. More precisely, because this model describes mesoscopic dynamics, we introduce a microscopic cutoff length $2\pi/k_c$, where the amplitude of the Fourier mode with $|\boldsymbol{k}| > k_c$ is set to zero.

Specifically, we study a system in which the mesoscopic free energy density $f(\phi)$ has two local minima at ϕ_1 and ϕ_2. We assume $\phi_1 < \phi_2$ without loss of generality. We impose periodic boundary conditions in the y direction and $\phi(\boldsymbol{r}) = \phi_1$ at $x = -L$ and $\phi(\boldsymbol{r}) = \phi_2$ at $x = L$. A one-dimensional planer interface is initially prepared at $x = 0$. We then observe the motion of the interface. The goal here is to determine the expectation value of the steady propagating velocity of the interface.

We first consider the case $T = 0$ with $f(\phi)$ fixed. The model given by (2) becomes a deterministic equation. The steady propagation solution $\phi_0(z)$ with $z = x - c_0 t$ satisfies

$$-c_0\partial_z\phi_0(z) = -\Gamma[f'(\phi_0) - \kappa\partial_z^2\phi_0], \quad (4)$$

where c_0 is the steady propagation velocity of the interface. Mathematically, (4) is a nonlinear eigenvalue problem for the solution ϕ_0 with a special value of c_0. Thus, $\phi_0(z)$ and c_0 are simultaneously determined. The explicit form of the solution is not generally written, but we can easily confirm the following relation in the limit $L \to \infty$ [10]:

$$c_0 = \Gamma_{\text{int}}[f(\phi_2) - f(\phi_1)], \quad (5)$$

with

$$\Gamma_{\text{int}} = \frac{\Gamma}{\int_{-\infty}^{\infty} dz[\partial_z\phi_0(z)]^2}. \quad (6)$$

The relation (5) indicates that the free energy density difference between the two local minima drives the interface to decrease the total free energy. The mobility of the interface is then given by (6).

When $T > 0$, the noise modifies the propagation velocity. To extract this effect clearly, we study the case $c_0 = 0$, which holds when $f(\phi_2) = f(\phi_1)$. We then consider the weak noise limit $T \to 0$ ignoring nucleation events in the bulk. Let $\theta(y, t)$ be the x coordinates of the interface at time t. The expectation of the fluctuating quantity $\theta(y, t)$ approaches a steady propagating state expressed as

$$\langle\theta(y, t)\rangle_{\text{ss}} = ct + \text{const}. \quad (7)$$

A finite value of c was reported for the case $d = 1$ [8], in which the nature of the driving force was found to be entropic. That is, when the fluctuation intensity around $\phi = \phi_1$ is larger than that around $\phi = \phi_2$, the entropy density in the region with $\phi = \phi_1$ is larger and then the region of $\phi = \phi_1$ becomes larger, leading to $c > 0$. The formula for c is expressed as [8]

$$c = \Gamma_{\text{int}}\frac{T}{2}\left(\frac{1}{\xi_2} - \frac{1}{\xi_1}\right) + O(T^{3/2}), \quad (8)$$

where $\xi_i \equiv \sqrt{\kappa/f''(\phi_i)}$ is the correlation length of fluctuations in the bulk region with $\phi = \phi_i$. It should be noted that (8) was confirmed by numerical simulations [8].

In this Letter, we study c for the system in two dimensions. Because the driving force of the interface motion is entropic, we expect that an entropic contribution $s(\phi)$ to the macroscopic free energy plays an essential role in the determination of c. We then conjecture

$$c = \Gamma_{\text{int}}\{-T[s(\phi_2) - s(\phi_1)]\} + O(T^{3/2}), \quad (9)$$

which means that the difference between the entropic contributions in each bulk region leads to the driving force. The question now is whether or not c can be expressed in such a form. Even if the form of (9) is correct, the functional form of $s(\phi)$ is not immediately obtained from the model (2). We thus need to derive c for the system in two dimensions. However, because the derivation method in Ref. [8], which follows the method proposed in Refs. [11–13], is specific to the one-dimensional case, we have to develop a general method of deriving c.

Main result.—We derive the stochastic interface dynamics from the stochastic model (2). As far as deterministic systems are concerned, there have been many methods used to derive the equation for interface motion [14–20]. The essence of these methods is to extract the interface motion as the slowest dynamics while separating other fast variables. We generalize the methods above to analyze stochastic systems in one and higher dimensions. We then obtain the formula (9) with

$$s(\phi_i) = -\frac{1}{2}\int_{|p|\le k_c}\frac{dp}{2\pi}\frac{\xi_i^{-2}-p^2}{p^2+\xi_i^{-2}}, \tag{10}$$

in one dimension and

$$s(\phi_i) = -\frac{1}{2}\int_{|\boldsymbol{p}|\le k_c}\frac{d^2\boldsymbol{p}}{(2\pi)^2}\frac{\xi_i^{-2}}{|\boldsymbol{p}|^2+\xi_i^{-2}}. \tag{11}$$

in two dimensions. The right-hand side of (10) is calculated as

$$s(\phi_i) = -\frac{1}{2\pi}\left[\frac{2}{\xi_i}\tan^{-1}(\xi_i k_c) - k_c\right]. \tag{12}$$

By substituting this result into (9) and taking the limit $k_c \to \infty$, we obtain (8). Then, for the two-dimensional case, the right-hand side of (11) is calculated as

$$s(\phi_i) = -\frac{1}{8\pi\xi_i^2}\ln(\xi_i^2 k_c^2 + 1), \tag{13}$$

where the cutoff wave number k_c should remain finite. This means that the stationary propagation velocity for the model with $d = 2$ singularly depends on the ultraviolet cutoff k_c. In other words, we need to specify a value of the cutoff k_c to study a measurement result of the propagating velocity.

Numerical simulations.—Because the cutoff dependence of the formula (13) is rather striking, we now confirm this result using numerical simulations. We note that $s(\phi_1) = s(\phi_2)$ when $f''(\phi_1) = f''(\phi_2)$. Therefore, an asymmetric landscape of $f(\phi)$ is necessary for the appearance of the entropic driving force. On the basis of this fact, we assume the local free energy density $f(\phi)$ is given by

$$f(\phi) = \left(\frac{1-\exp[b_1(\phi-1)]}{1-\exp[b_1(\phi_0-1)]}\frac{1-\exp[-b_2(\phi+1)]}{1-\exp[-b_2(\phi_0+1)]}\right)^2, \tag{14}$$

as in Ref. [8]. Here, f satisfies the condition that $\phi_1 = -1$, $\phi_2 = 1$, and $f(\phi_1) = f(\phi_2)$. In Fig. 1, we show the form of the local free energy density f. It can be seen that the potential is highly asymmetric, i.e., $f''(\phi_1) \gg f''(\phi_2)$. Examples of asymmetric free energy density f were presented in Refs. [21,22].

We define a discrete model on a square lattice with a spatial mesh size of Δx, considering that Δx should be smaller than ξ_1 and ξ_2. The discrete model is obtained by discretizing (2), where $\partial_x^2 + \partial_y^2$ is replaced by the finite difference Laplacian. For the initial condition

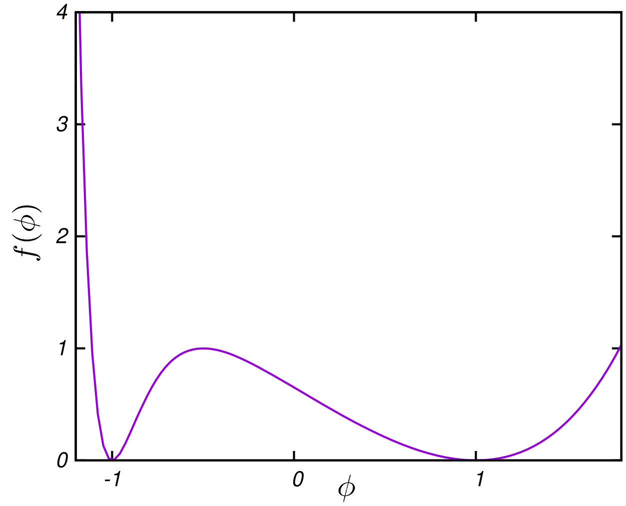

FIG. 1. Shape of the mesoscopic free energy density (14) with $b_1 = 0.5$, $b_2 = 5.0$, and $\phi_0 = -0.5$.

$$\phi(\boldsymbol{r}, 0) = \frac{\phi_2 - \phi_1}{2}\tanh\left(\frac{x}{40}\right) + \frac{\phi_1 + \phi_2}{2}, \tag{15}$$

the stochastic time evolution is performed using the Heun method. We then measure

$$\Phi(x, t) \equiv \frac{1}{L_y}\int_0^{L_y} dy\phi(x, y, t), \tag{16}$$

which describes the x profile averaged in the y direction. In Fig. 2, we show an example of $\Phi(x, t)$ for several values of t. For the interface position $X(t)$ defined by $\Phi(X(t), t) = 0$,

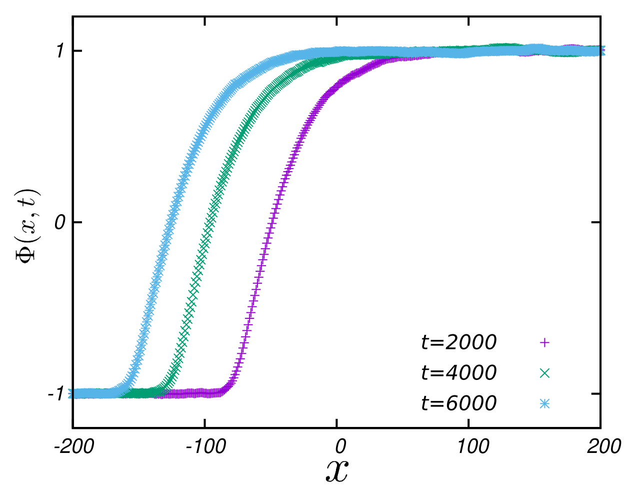

FIG. 2. Time evolution of the pattern averaged in the y direction. The free energy density $f(\phi)$ is the same as that in Fig. 1. The other parameter values are $\kappa = 1600$, $\Gamma = 0.1$, $T = 0.5$, $L = 400$, and $L_y = 100$. Noting that $\xi_1 = 4.33$ and $\xi_2 = 27.3$ for these parameters, we choose $\Delta x = 1.0$ and $\Delta t = 5.0 \times 10^{-4}$ [23].

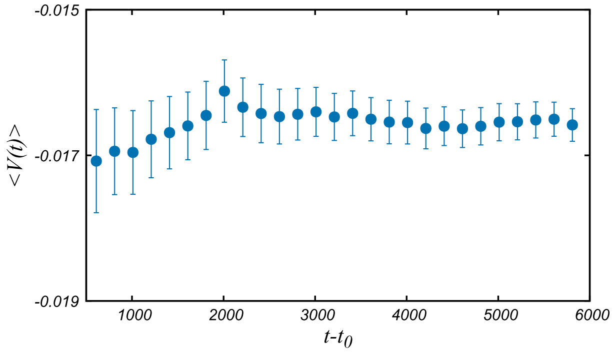

FIG. 3. $\langle V(t)\rangle$ as a function of $t - t_0$. The parameter values are the same as those in Fig. 2. $t_0 = 50$. Eighty samples are used to estimate $\langle V(t)\rangle$ with error bars.

we define a time-averaged velocity as

$$V(t) \equiv \frac{X(t) - X(t_0)}{t - t_0}, \tag{17}$$

where t_0 is chosen to be much larger than the relaxation time to the steady propagating state. In Fig. 3, we plot the expectation value of $V(t)$, which gives the numerically estimated value of c. We then confirm that c is proportional to T for $T \leq 1$ [23].

We now perform the same calculation for systems with different values of Δx. The results are displayed in Fig. 4. It is observed that c does not go to a definite value as Δx becomes smaller under the condition that $\Delta x < \xi_1 \ll \xi_2$, which is in contrast with the one-dimensional case. To compare the numerical data with the theoretical result, we overlay the graph of (9) with (13) in Fig. 4, where we choose

$$k_c = \sqrt{\left(\frac{2\pi}{\Delta x}\right)^2 + \left(\frac{2\pi}{\Delta x}\right)^2} = \frac{2\sqrt{2}\pi}{\Delta x}, \tag{18}$$

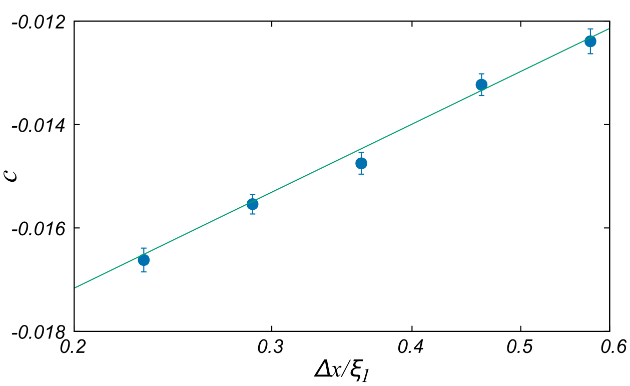

FIG. 4. Δx dependence of the propagating velocity c for the system in two dimensions. The parameter values are the same as those in Fig. 2. The blue circles represent numerical results given by $\langle V(t = 6000)\rangle$. The green curve represents (9) with (13). We checked the validity of the values of the numerical parameters Δx and Δt [23].

which corresponds to the largest magnitude of the wave vector in the numerical simulations. We find that the theoretical calculation is consistent with the numerical simulations. We thus conjecture that $|c|$ diverges in the limit $\Delta x \to 0$ even in numerical simulations.

Sketch of the derivation.—We first take the limit $L \to \infty$. Let T be sufficiently small such that the noise effect can be studied as a perturbation of the deterministic system. To express this smallness, we replace T with $\epsilon^2 T$, where ϵ is a small dimensionless parameter. When a perturbation is imposed on the stationary solution $\phi_0(x)$, the response is divided into the interface motion $\theta(y, t)$ and the rest. That is, we express the perturbation solution as

$$\phi(x, y, t) = \phi_0(z) + \epsilon\rho_1(z, y, t) + O(\epsilon^2), \tag{19}$$

with the comoving coordinate $z = x - \theta(y, t)$. Assuming that $\partial_y\theta$ is small and proportional to ϵ, we introduce a large scaled coordinate Y as $Y = \epsilon y$, and define $\Theta(Y, t) = \theta(y, t)$. The time evolution of $\Theta(Y, t)$ is described by

$$\partial_t\Theta = \epsilon\Omega_1([\Theta]) + \epsilon^2\Omega_2([\Theta]) + O(\epsilon^3), \tag{20}$$

where $[\Theta]$ represents the dependence of $\partial_Y\Theta$ and $\partial_Y^2\Theta$. By substituting (19) and (20) into (2), we can determine the statistical properties of ρ_1, Ω_1, and Ω_2. This calculation method is regarded as a generalization of the method in Ref. [14] to stochastic systems.

The stationary propagation velocity c is obtained as $c = \epsilon\langle\Omega_1\rangle_{ss} + \epsilon^2\langle\Omega_2\rangle_{ss} + O(\epsilon^3)$. We calculate $\langle\Omega_1\rangle_{ss} = 0$ and

$$\langle\Omega_2\rangle_{ss} = \frac{\Gamma}{2}\frac{\int_{-\infty}^{\infty} dz(\partial_z\phi_0)f^{(3)}(\phi_0)\langle\rho_1^2\rangle_{ss}}{\int_{-\infty}^{\infty} dz(\partial_z\phi_0)^2}. \tag{21}$$

This formula was derived in Ref. [27]. Now, from the time-reversal symmetry of the steady state in the comoving frame, we find that the integral of the numerator in (21) is expressed as $\lim_{\Lambda\to\infty}[\Psi(z = \Lambda) - \Psi(z = -\Lambda)]$ using a function $\Psi(z)$ given by

$$\begin{aligned}\Psi(z) &= f^{(2)}(\phi_0(z))\langle\rho_1(z, y)^2\rangle_{ss} \\ &\quad - \kappa\left[\langle(\partial_z\rho_1(z, y))^2\rangle_{ss} - \left\langle(\partial_y\rho_1(z, y))^2\right\rangle_{ss}\right].\end{aligned} \tag{22}$$

Therefore, (21) takes the form (9) with

$$Ts(\phi_i) = -\frac{1}{2}\lim_{\Lambda\to\infty}\Psi(\mu_i\Lambda), \tag{23}$$

where $\mu_1 = -1$ and $\mu_2 = 1$. Finally, by evaluating $\Psi(z)$, we obtain (11). The calculation result is immediately generalized to d-dimensional systems [23]. In particular, for the case $d = 1$, we obtain (10).

Concluding remarks.—We have derived the formula (9) with (11) for the entropic driving force in the mesoscopic model (2). Although we have studied the case $c_0 = 0$, it is straightforward to derive the propagation velocity for $c_0 \neq 0$ as $c_0 + c$, which is denoted by c_*. We have found that the entropic force singularly depends on the microscopic cutoff of the mesoscopic model (2) in two dimensions. This discovery suggests the need for further study.

The most important challenge is an experimental observation of the entropic force in two or three dimensions. As an experimental system, we propose to study a spin-crossover complex where high-spin and low-spin states can coexist with an interface [9]. Since curvatures of the free energy at these states are different, we expect the entropic contribution of the force generating the interface propagation. Here, through the measurement of the space-time correlation of order parameter fluctuations, Γ and the correlation lengths are evaluated. By measuring c_* directly at sufficiently low temperatures T_1 and T_2, we may estimate the value of k_c by using $c_*(T_2) - c_*(T_1) \simeq -\Gamma_{\rm int}(T_2 - T_1)(s(\phi_2) - s(\phi_1))$ with the assumption that in the low temperature region $f(\phi)$ does not depend on T.

From the theoretical viewpoint, it is significant to generalize our formula (9) for studying various systems such as conserving systems [28] and out-of-equilibrium systems [29–31]. More fundamentally, in pursuit of a microscopic understanding of the cutoff, one can attempt to consider the derivation of the mesoscopic description from more microscopic systems such as lattice models [32] or Hamiltonian systems [33]. Although there have been many related studies since Ref. [34], the explicit determination of coefficients of the mesoscopic model is not easy as argued in Ref. [35]. To develop a theory explaining the cutoff dependence based on a microscopic description should be another goal of nonequilibrium statistical mechanics. As another direction of study, the universality class of stochastic interface motion will be explored by studying the fluctuation properties of the interface motion. With regard to this problem, we point out that the microscopic cutoff dependence observed in the Kardar-Parisi-Zhang equation [25,26] comes from a nonlinear term that is not relevant in our problem [23]. Thus, the microscopic cutoff dependence reported in this Letter has never been studied so far.

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