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Nonequilibrium Temperature and Fluctuation Theorem in Soft-Mode Turbulence

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Abstract

Non-thermal Brownian motion of a particle in soft-mode turbulence (SMT) in the electroconvection of a nematic liquid crystal has been experimentally investigated to clarify the statistical and thermodynamical aspects of SMT using the Lagrangian picture in hydrodynamics. The effective temperature for SMT is obtained in two different ways based on the definition of the diffusion coefficients due to non-thermal particle fluctuations: the Einstein relation and the fluctuation theorem. The temperatures from both methods agree well and exhibit a high value of 10^6 K. They depend on the coarse-graining time, which reflects the anomalous properties of the macroscopic fluctuations in the SMT.

Key words: soft-mode turbulence, fluctuation theorem, Einstein relation, electroconvection

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1. Introduction

Spatiotemporal chaos (STC) has been investigated as one of the most important phenomena in nonlinear dynamics in spatially extended nonequilibrium systems (1). To date, most experimental and theoretical research into STC has been performed under the Eulerian picture, that is, with the pattern dynamics described by field variables. The Lagrangian picture, on the other hand, which focuses on particle motion, gives other and/or additional information about the fluid motion and properties in STC (2; 3). The Lagrangian picture is often more important than the Eulerian picture from the viewpoint of statistical mechanics, because it is directly related to transport phenomena.

Brownian motion is a well-known thermal fluctuation near thermal equilibrium which is conventionally described by the linear Langevin equation and is directly connected to various transport coefficients via the fluctuation-dissipation theorem. The treatment of thermal Brownian motion near equilibrium may be extended to non-thermal Brownian motion in nonlinear systems far from equilibrium, such as STC (4; 5). Previous research (4; 6) has discussed the statistical mechanical properties of soft-mode turbulence (SMT), a kind of STC, in electroconvection of a nematic liquid crystal, via non-thermal Brownian motion of particles by SMT (7; 8). The macroscopic Langevin equation has been proposed for the non-thermal Brownian motion (4):

$$m\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{v}(t) = -\Gamma\mathbf{v}(t) + \boldsymbol{\Xi}(t). \tag{1}$$

Here, m and $\mathbf{v}(t) = \mathrm{d}\mathbf{r}(t)/\mathrm{d}t$ are the mass and velocity of the particle, respectively. $\Xi(t)$ is the random force caused by the SMT. The lag between $\mathbf{v}(t)$ and the local flow velocity at the position $\mathbf{r}(t)$ of the particle is due to the finite mass effect (9). Therefore, the friction coefficient Γ includes both macroscopic (SMT) and microscopic (thermal fluctuations) information. If the non-thermal Brownian motion is described by Eq. (1), we have to separate Γ and the random force $\Xi(t)$ from the time series of $\mathbf{r}(t)$.

In the present research, we have measured the non-thermal Brownian motion under a constant and additional external force in Eq. (1). Based on the response to the external force, a Langevin-type description in the Lagrangian picture of the SMT could be developed. Furthermore, based on the fluctuation theorem (10) , the non-thermal Brownian motion has been discussed. Consequently, we experimentally obtain the effective temperature characterizing the strength of the macroscopic fluctuations.

2. Experiment

2.1. Electrohydrodynamics and non-thermal Brownian motion in SMT

SMT is observed in the homeotropic alignment systems of nematic liquid crystals (8). The nematic director beyond the Fréedericksz transition point becomes a Nambu–Goldstone mode. Then, the nonlinear coupling between the Nambu–Goldstone mode and the electroconvective mode beyond the convective threshold V_c of the applied voltage induces the SMT (see Ref. (7)).

Usually, in a spatially extended nonlinear system far from equilibrium, a regular structure appears first beyond a primary threshold. As the control parameter of the system increases, it becomes unstable and STC appears beyond a secondary threshold. In homeotropic systems, however, SMT directly occurs at the primary threshold via a single supercritical bifurcation (7). Macroscopic fluctuation such as SMT is a phenomenon in the weakly nonlinear regime. Therefore, the study of SMT is expected to provide a new perspective on macroscopic nonlinear fluctuations due to the smooth connection between SMT and linear states.

2.2. Preparation and motion of a particle

In order to trace the random motion of a particle in SMT, we prepared an experimental system with a small particle in a standard cell in homeotropic alignment (4). The diameter and density of the particle (silica bead) were $a = 3.0$ μm and $\rho = 2.2 \times 10^3$ kg/m^3, respectively. A circular transparent electrode of diameter 13.0 mm was used and the thickness of the sample was 94 μm. The dielectric constant and electric conductivity parallel to the director were $\epsilon_{||} = 4.0$ and $\sigma_{||} = 5.3 \times 10^{-7}\Omega^{-1}\mathrm{m}^{-1}$, respectively.

In typical experiments of electroconvection in nematic liquid crystals, a convective structure is observed as a two-dimensional pattern in the x–y plane. We focus therefore on non-thermal Brownian motion in the x–y plane. The SMT occurs in the weakly nonlinear regime and the direction of the convective rolls spatiotemporally fluctuates in regular convective roll structures. In previous research, samples was placed on a stage of a microscope and observed from above. In the present research, in contrast, in order to use gravity as an additional external force, we placed the sample vertically and observed from the side using a stereomicroscope (Leica MZ7). As a result, the particles were subjected to the following constant force in the y-direction:

$$F = (\rho - \rho_0)\frac{4}{3}\pi a^3 g = 1.3 \text{ pN}. \quad (2)$$

Here, $\rho_0 = 1.02 \times 10^3$ kg/m^3 is the density of the nematic liquid crystal (MBBA) and g is the gravity acceleration. Consequently, we may consider the following macroscopic Langevin equation to describe the particle motion:

$$m\frac{\mathrm{d}}{\mathrm{d}t}v_y(t) = -\Gamma v_y(t) + \Xi_y(t) + F, \quad (3)$$

where $v_y(t)$ and $\Xi_y(t)$ are the y-components of $\mathbf{v}(t)$ and $\boldsymbol{\Xi}(t)$, respectively.

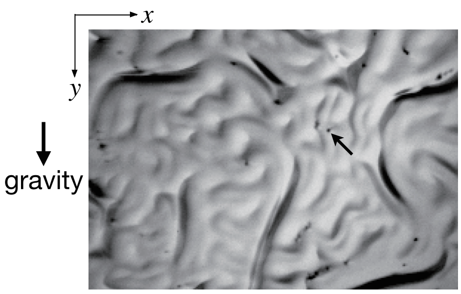

Figure 1: A particle in soft-mode turbulence. We traced the position $\mathbf{r}(t) = (x(t), y(t))$ of the particle indicated by the arrow.

In the oblique rolls regime (11), the fluctuations of the SMT are isotropic and strong enough to drive the particles (12). In order to perform the experiments in the oblique rolls regime, we set the frequency f of the applied voltage to $f = 110$ Hz $< f_\mathrm{L} \simeq 1500$ Hz, where f_L is the Lifshitz frequency (11).

Images containing the particles were sampled at a time interval of 1 s over 300 s using a charge coupled device camera (Retiga 2000R). The time series of the position $\mathbf{r}(t) = (x(t), y(t))$ of a particle was recorded from the images using a custom-written version-up program in ImageJ. In order to enhance the statistical precision, we analyzed 22–37 time series for each ε.

3. Results and Discussion

3.1. Assumption of the Einstein Relation

By taking a long-time average of Eq. (3), the following relation is obtained:

$$\Gamma = \frac{F}{\overline{v_y}}, \tag{4}$$

since $\overline{\Xi(t)} = 0$, where $\overline{\cdots}$ represents the long-time average of $\cdots$. Γ is recognized as the mobility for F. We can experimentally obtain values of Γ from the experimental data of $\overline{v_y} = \left(y(t_{\mathrm{f}}) - y(0)\right)/t_{\mathrm{f}}$ where $t_{\mathrm{f}} = 300$ s is the observation time.

In previous research, we presented the time-scale dependence of the diffusion coefficients in the absence of externally deterministic forces (6), as

$$D_{\mathrm{s}}(\tau) = \frac{\langle |\mathbf{r}(t+\tau) - \mathbf{r}(t)|^2 \rangle}{2n\tau}, \tag{5}$$

in order to discuss the properties of diffusion in SMT. Here, n is the space dimension of the system. $D_{\mathrm{s}}(\tau)$ in SMT shows an anomalous dependence on τ smaller than $\tau_{\mathrm{c}} \sim 1000$ s, the anomalous diffusion and the value of $D_{\mathrm{s}}(\tau)$ monotonically increases proportional to $\tau^{0.32}$. For $\tau > \tau_{\mathrm{c}} \sim 1000$ s, $D_{\mathrm{s}}(\tau)$ becomes normal diffusion, i.e. constant in τ (6; 8). Thus, without any externally deterministic force, the diffusion process in SMT has already been

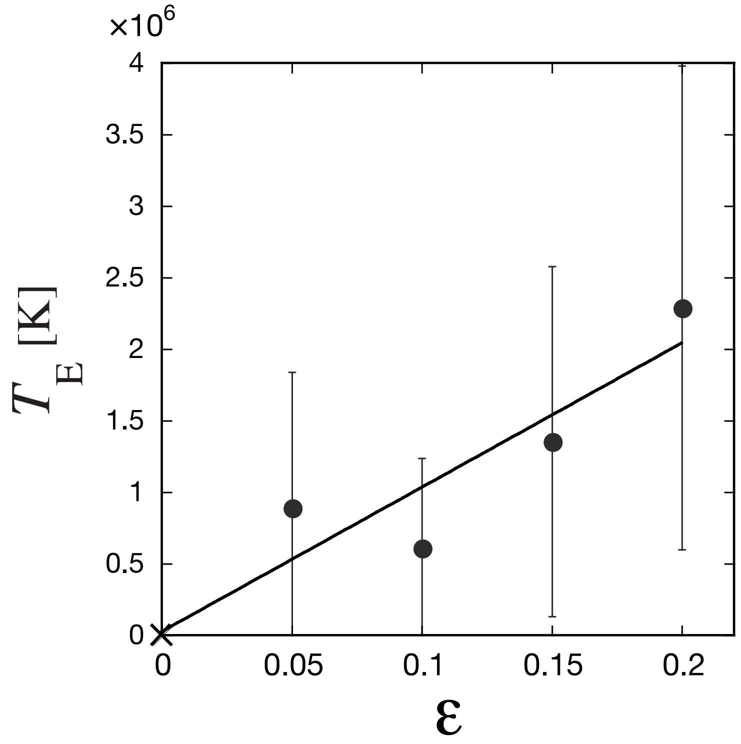

Figure 2: ε-dependence of $T_{\rm E}(t_{\rm f})$. Values averaged from the 22–37 time series are plotted and error bars are drawn for the standard deviation. We believe that the large errors are due to anomalous diffusion, since $t_{\rm f}$ is smaller than $\tau_{\rm c}$. However, more detailed studies are required to confirm our beliefs and allow for more sophisticated discussions. At $\varepsilon = 0$, the ambient temperature of the experimental system is plotted as a cross.

well studied. Therefore, we focus here on the diffusion in the y-direction, the direction in which the external force F is applied. The diffusion coefficient is now defined as

$$D'_{\rm s}(\tau) = \frac{\langle |Y(t+\tau) - Y(t)|^2 \rangle}{2\tau}, \tag{6}$$

where $Y(t) = y(t) - \overline{v_y}\, t$.

We assume the Einstein relation

$$T_{\rm E}(\tau) = \frac{\Gamma D'_{\rm s}(\tau)}{k_{\rm B}}, \tag{7}$$

where $k_{\rm B}$ is the Boltzmann constant. The "temperature" $T_{\rm E}(\tau)$ quantitatively characterizes the macroscopic fluctuations in the SMT. We can recognize the effective temperature $T_{\rm E}(\tau)$ as the average energy of the force applied to the particle by the SMT. For $T_{\rm E} = 10^6$ K the fluctuation of the SMT is quite macroscopic and strong. In Fig. 2, the ε-dependence of $T_{\rm E}(t_{\rm f})$ is shown. The strength of the fluctuations in the SMT corresponds to $O(10^6)$ K in terms of the temperature defined here. It is thought that the linear relation between ε and $T_{\rm E}(t_{\rm f})$ is consistent with the scaling relations of the correlation time $\propto \varepsilon^{-1}$ (7) and the correlation length $\propto \varepsilon^{-1/2}$ (13) for the SMT.

3.2. Fluctuation Theorem

The fluctuation-dissipation theorem is the most useful tool for understanding the behavior of a linear system near thermal equilibrium. However, a generalized fluctuation-dissipation theorem including nonlinear systems far from equilibrium has not been established. In 1995, a new theorem for fluctuations, generalized to include nonlinear systems, was presented (10) and the experimental verification and theoretical proof of this theorem have been well studied (14; 15; 16). Though the theorem is expected to be the counterpart of the fluctuation-dissipation theorem, its practical application has not yet been presented. In the present research, we experimentally obtain a parameter which quantitatively characterizes STC from the Lagrangian picture by using the fluctuation theorem (17; 18).

In the present system, the external force F gives a particle some work. Though the work averaged over a long time may be positive, it can take negative values at instantaneous steps. Therefore the mean power during τ, given by

$$w_\tau(t) = \frac{F \cdot s_\tau(t)}{\tau}, \tag{8}$$

where $s_\tau(t) = y(t+\tau) - y(t)$, fluctuates in time, and the probability distribution function $P(w_\tau)$ of w_τ depends on the coarse-graining time τ. The fluctuation theorem (16)

$$\frac{P(w_\tau)}{P(-w_\tau)} = \exp\left(\frac{w_\tau}{\Theta}\tau\right) \tag{9}$$

has a quantitative relation with $P(w_\tau)$. Θ corresponds to the energy of the random force which drives a particle. In the case of thermal Brownian motion, $\Theta = k_{\mathrm{B}}T$ where T is the temperature of the heat bath.

In this subsection, ε was fixed at 0.1, and we show the averages of 29 time series of $\mathbf{r}(t)$. As shown in Fig. 3, $P(w_\tau)$ is Gaussian [2] and its form depends on τ. From the distribution functions,

$$\Phi(w_\tau) = \ln\left[\frac{P(w_\tau)}{P(-w_\tau)}\right] \tag{10}$$

[2] The form of a distribution function may depend on the weight and the size of the particle as well as ε (6). The detail will be reported elsewhere.

can be calculated. Figure 4 shows $\Phi(w_\tau)$ for several values of τ. Though the linear relation between $\Phi(w_\tau)$ and w_τ is seen, there is scatter in the data points. If the distribution function is completely Gaussian, there is no scatter in $\Phi(w_\tau)$. Actually, the distribution functions in Fig. 3 show small deviations from the Gaussian function. The scatter in Fig. 4 comes from these deviations. However, it can be recognized that $P(w_\tau)$ in Fig. 3 is essentially Gaussian. Therefore, we can conclude that the fluctuation theorem holds for non-thermal Brownian motion in SMT.

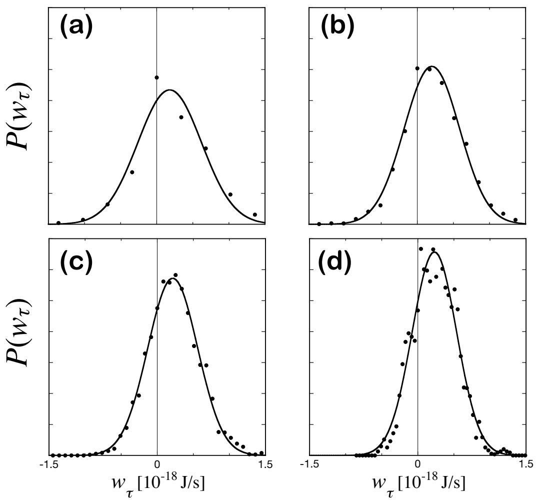

Figure 3: $P(w_\tau)$. (a) $\tau = 10$ s. (b) $\tau = 20$ s. (c) $\tau = 40$ s. (d) $\tau = 80$ s.

From Eq. (9), the slopes α_τ of $\Phi(w_\tau)$ in Fig. 4 should be equal to τ/Θ. Therefore, we can also obtain the effective "temperature" due to macroscopic fluctuations in a different way from T_E, as

$$T_\mathrm{FT}(\tau) = \frac{\tau}{k_\mathrm{B}\,\alpha_\tau}. \tag{11}$$

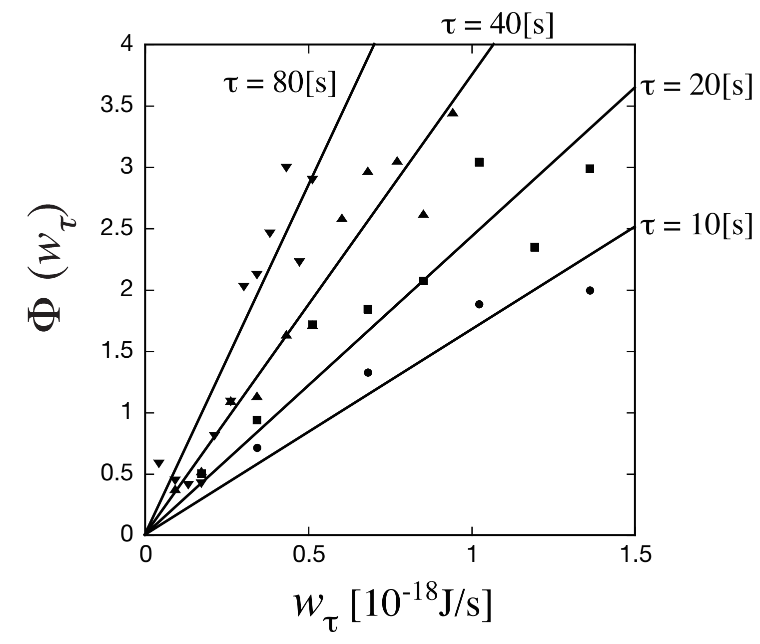

Figure 4: $\Phi(w_\tau)$.

Figure 5 shows the τ-dependences of T_E and T_FT.

This result means that the strength of the fluctuations in SMT depends on the coarse-graining time τ, similar to the diffusion coefficient D_s (6). Both the effective temperatures T_E and T_FT show good agreement below $\tau \sim 200$ s.

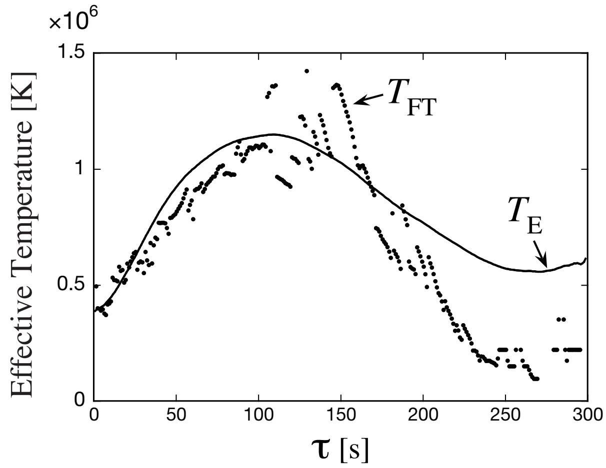

Figure 5: τ-dependences of the effective temperatures T_E and T_FT.

However, the difference between the temperatures becomes large beyond $\tau \sim 200$ s. Both decrease for $\tau \gtrsim 100$–150 s. As described for the previous result, $D_\mathrm{s}(\tau)$ monotonically increases below $\tau_\mathrm{c} \sim 1000$ s (6). According to the result, T_E is expected to increase monotonically for $\tau < \tau_\mathrm{c}$. As mentioned in Section 1, Γ includes not only macroscopic effects of SMT, but also microscopic friction induced by the lag between the velocity of the particle and local flow velocity. In the present experiments, since the particles were dragged by a constant force, the effect of the lag in the y-direction may be stronger than sthat in the x-direction, which may manifest as strange behavior in the y-direction. Thus, a sufficiently long time measurement may be necessary to clarify the fine structures observed in Fig. 5.

4. Summary

We have experimentally investigated non-thermal Brownian motion under gravitational acceleration as a constant force in SMT, i.e. STC observed in an electroconvective system of a nematic liquid crystal. A particle doped in the system shows a random motion due to the fluctuations of the SMT. This non-thermal Brownian motion reveals statistical properties and information on SMT in the Lagrangian picture. We have presented two different ways to determine the effective temperature of SMT containing such information of macroscopic random motions.

By measuring the long time average of the velocity of the particle, the macroscopic friction coefficient was obtained. By applying the Einstein relation to the present non-thermal Brownian motion, the effective temperature T_{E} was obtained from the friction coefficient and the diffusion coefficient.

The other effective temperature T_{FT} was obtained by using the fluctuation theorem. Our observation w_τ is a typical response to the external force F and distributes around the average by kicks from the fluctuation of the SMT.

Both effective temperatures show good agreement for small coarse-graining time. It should be stressed that both effective temperatures show dependence on the coarse-graining time. This property may appear also in other nonequilibrium open systems, because it is due to anomalous transport of the dissipative structures (4; 6). The effective temperatures are useful for describing statistical mechanical properties of matter transport in STC.

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