Correlation Functions in Homeotropic Nematics: Non-Exponential Relaxation of Soft-Mode Turbulence

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1. Introduction

Convection is an experimentally-obtained system of steady-state nonequilibrium physics. The Rayleigh– Benard convection is a convection caused by heat transfer. Another example is electroconvection observed in nematic liquid crystals, in which the control parameter is the ac voltage applied to system. The characteristic length of the electroconvection is much shorter than that of the Rayleigh–Benard convection, and consequently characteristic time scales of the electroconvection are accessible experimentally. Thus, electrohydrodynamic convection is one of the most convenient phenomena to investigate pattern formation of the nonequilibrium physics.

There are some types of layer alignment in nematic liquid crystal; one of them is planar alignment, where the director aligns parallel to substrates (x-direction), and another is homeotropic alignment, where the director aligns perpendicular to substrates (z-direction). The rubbing to x-direction on the substrates' surface (x-y plane) is treated to make planer systems, and intrinsically breaks the rotational symmetry. With increasing magnitude V and fixed frequency f of the applied ac voltage, static convection patterns appear above a threshold voltage [1, 2]. On the other hand, in the homeotropic systems, the rotational symmetry on x-y plane is alive at a sufficiently low voltage. As the applied voltage increases, the Freédericksz transition [3] occurs at a threshold voltage V_F and breaks the rotational symmetry spontaneously, accompanied by the Nambu–Goldstone mode at zero wave number [4-6]. With further increasing the voltage, a secondary transition occurs at another threshold voltage V_c and the electroconvection occurs. Consequently, a spatially and temporally complex dynamics is generated by nonlinear interaction between the Nambu–Goldstone mode and the convection mode that corresponds to nonzero wave number [7-9]. This can be regarded as experimentally-obtained spatiotemporal chaos, and was named soft-mode turbulence (SMT) because of the softening of the state's macroscopic fluctuations [7].

We investigated SMT dynamics by measuring temporal autocorrelation functions of turbulence-like pattern. It had long been considered that the relaxation was described by the simple exponential. However, we revealed that the relaxation deviates near the threshold of electroconvection [10]; it is well-fitted by the compressed exponential, which describes the dynamics of jammed systems. It suggests a similarity between dynamics of SMT and that of glass-forming liquids (GFL). The compressed exponential relaxation in GFL arises from cooperatively rearranging; indeed, spatially and temporally fluctuating domains have been experimentally observed near the glass transition point. On the other hand, in SMT, there exist patch domains where electroconvective rolls are parallel while the pattern is disordered as a whole. Thus, the domain of SMT corresponds to the cooperatively rearranging region of GFL, and the coherency in the patch domains leads to the non-exponential relaxation in SMT.

Our previous study has been focused on the *total* autocorrelation function including entire wave-number information, while the wave-number dependence has been discussed in the context of GFL. The temporal correlations of each wave number, i.e., *modal* autocorrelation functions, defined by

$$\hat{U}_k(t) = \langle u_k(t) u_k^*(0) \rangle P_k^{-1},$$
(1)

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are more suitable for studying SMT dynamics, where $u_k(t)$ denotes the Fourier transform of the transmitted light intensity and P_k the spatial power spectrum. In this international conference, we aim to present our experimental results of the modal correlation function and discuss an interesting phenomenon found in SMT.

2. Experimental setup

We study a two-dimensional pattern dynamics of SMT observed in the homeotropic alignment of nematic liquid crystals [7]. The space between two parallel glass plates was filled with MBBA [N-(4–Methoxybenzilidene)–4– buthylaniline]. The thickness between the plates is 27 μ m. The surfaces were laid by DMOAP (N, N-dimethyl–N- octadecyl–3–aminopropyl-trimethoxysilyl chloride 50%) to trigger the homeotropic alignment, and were coated with indium tin oxide (ITO) which is a circular electrode with the radius 1.3 cm. The dielectric constant parallel to the director was $\epsilon_{\parallel} = 6.25$ and the electric conductivity parallel to the director $\sigma_{\parallel} = 1.17 \times 10^{-7} \ \Omega^{-1}$ m⁻¹. Note that the dielectric constant anisotropy $\epsilon_a := \epsilon_{\parallel} - \epsilon_{\perp}$ is negative.

An ac voltage $V(t) = \sqrt{2}V \cos(2\pi ft)$ was applied to the sample. A normalized voltage $\varepsilon = (V/V_c)^2 - 1$ was employed as a control parameter, where V_c was 7.78 ± 0.05 V. Two types of SMT pattern arise; oblique rolls in $f < f_L$ and normal rolls in $f > f_L$, where f_L denotes the Lifshitz



Figure 1: A typical snapshot of the SMT with the white scale bar indicating 100 μ m, where the control parameters are $\varepsilon = 0.1$.

frequency [7, 8]. In this study, we fix f = 100 Hz that is less than f_L . The temperature is controlled at 30.00 ± 0.05 °C. Before each data sampling, we waited for 10 min at $V_w = 6.0$ V and then for 10 min at desired V, where $V_F < V_w < V_c < V$.

The electroconvection pattern was observed by a microscope and was captured by a high-speed camera. In the region of the control parameters we set, SMT was successfully observed. A typical two-dimensional image is shown in Fig. 1, where the measurement area was $830 \times 830 \ \mu\text{m}^2$ ($450 \times 450 \ \text{pixels}$).

The power spectrum P_k obtained experimentally has a clear peak around $k_{\text{peak}} \simeq 0.321 \ \mu\text{m}^{-1}$. Its wavelength $\lambda_{\text{peak}} = 2\pi/k_{\text{peak}}$ is 19.6 μm corresponding to the fundamental period of electroconvectional rolls. We employ k_{peak} as the reference value; namely, the wave number is normalized as $\hat{k} := k/k_{\text{peak}}$.

3. Theory

Using the nonlinear projection formalism [11, 12], one can derive the evolutional equation renormalizing nonlinear fluctuations as

$$\frac{\mathrm{d}U_{ij}(t)}{\mathrm{d}t} = \mathrm{i}\omega_{il}U_{lj}(t) - \int_0^t \mathrm{d}s\Gamma'_{il}(t-s)U_{lj}(s),\tag{2}$$

where $i = \sqrt{-1}$, ω_{ij} denotes the mechanical coefficient, $U_{ij}(t)$ the temporal correlation function of the gross variables $A_i(t)$ and $A_j(0)$, and $\Gamma'_{ij}(t)$ denotes the memory function containing nonlinear fluctuations.

When the characteristic time scale of the microscopic fluctuations is much fast, $\Gamma'_{ij}(t)$ can be separated as

$$\Gamma'_{ij}(t) = 2\gamma^{(0)}_{ij}\delta(t) + \Gamma_{ij}(t), \qquad (3)$$

where $\gamma_{ij}^{(0)}$ denotes the bare friction coefficient and $\Gamma_{ij}(t)$ is represented by the correlation of the nonlinear fluctuations.

We here presume $\{u_k\}$ as a complete set of the gross variables in SMT. The evolution equation of the normalized modal time-correlation function is thus represented as

$$\frac{\mathrm{d}\hat{U}_k(t)}{\mathrm{d}t} = -\int_0^t \mathrm{d}s\Gamma'_k(t-s)\hat{U}_k(s).$$
(4)

Note that the mechanical coefficient vanishes analytically. If the assumption that the characteristic time is fast for the microscopic fluctuations holds even in SMT, we obtain

$$\frac{\mathrm{d}\hat{U}_{k}(t)}{\mathrm{d}t} = -\gamma_{k}^{(0)}\hat{U}_{k}(t) - \int_{0}^{t}\mathrm{d}s\Gamma_{k}(t-s)\hat{U}_{k}(s)$$
(5)

with

$$\Gamma'_k(t) = 2\gamma_k^{(0)}\delta(t) + \Gamma_k(t).$$
(6)

We can numerically obtain the memory function $\Gamma_k(t)$ from Eqn. (5) using the experimentally-obtained $\hat{U}_k(t)$.

4. Result and discussion

We have clarified that the memory effect due to nonlinear fluctuations has a finite time scale, implying the memory plays a important role for the pattern dynamics in SMT. Consequently, the modal relaxations have dual structure due to chaotic mixing [13]; the relaxation in the faster time regime is dominated by the algebraic (i.e., time-reversible) function

$$\hat{U}_k(t) = \frac{1}{1 + \left(t/\tau_k^{(a)}\right)^2},$$
(7)

and that in the slower time regime is dominated by the simple exponential (i.e., time-irreversible) one

$$\hat{U}_k(t) \propto \exp\left[-t \left/ \tau_k^{(e)} \right],$$
 (8)

where $\tau_k^{(a)}$ and $\tau_k^{(e)}$ denote the characteristic timescales for faster and slower regimes, respectively. Figure 2 shows the dual structure schematically for a specific parameter. Note that for the other parameters we found the dual structure as well. The dual structure has been theoretically predicted to date; however, this study has observed it experimentally for the first time.



Figure 2: Dual structure in a modal relaxation $\hat{U}_k(t)$ with $\hat{k} = 1.0$ and $\varepsilon = 0.1$. Open circle indicates experimental result, gray solid line the algebraic decay, and black dashed line the simple exponential decay.

In the oral presentation, we show our experimental results and discuss the dual structure of SMT in detail.

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