Characteristic Exponent of Normal and Oblique Rolls in Homeotropically Aligned Nematic Liquid Crystal

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Abstract. Soft-mode turbulence (SMT) is one of an experimental example of spatiotemporal chaos, observed in electroconvection system of homeotropically aligned nematic liquid crystal (NLC), due to a non-linear interaction between Nambu-Goldstone mode denoted by the $C(r)$-director and the convective mode $q(r)$. There are two types of stripe patterns in the SMT, namely normal rolls (NR) and oblique rolls (OR) which separated by a point of applied frequency, called the Lifshitz frequency ($f_L$). We report a study of the phase transition from normal to oblique rolls by observing the patterns with an applied frequency below and beyond of $f_L$. The temporal fluctuations of the pattern images had been analyzed using autocorrelation function. It fits with Kohlrausch Williams Watts (KWW) function, showing there is a dynamical glass-forming liquid in the transition of NR-OR regime. Also, we found a new type of defect in the NR regime which never been reported before, a dynamic defect which takes the shape of a ring first to a spot in the end.

1. INTRODUCTION
The changing of natural phenomenon that occurs around us is an extremely unstable system. A system in far from equilibrium which infinitely rich in a change of the dynamics sometimes exhibits a variety of pattern structure [1]. The most well-known phenomenon that triggers a pattern formation is turbulence in the fluid system. Recently, significant development related to the detailed classification of turbulence has been obtained; one of them is spatiotemporal chaos (STC) which are spatially and temporally disordered structure [2]. Very recently, one of the experimental models of STC that attracts a good deal of researcher’s attention is Soft-Mode Turbulence (SMT) which shows a special scenario for the transition to turbulence, because it directly appears from a quiescent state via single supercritical bifurcation [3].

The SMT has been found in electroconvection system of homeotropically aligned nematic liquid crystal (NLC). In the homeotropic alignment, the director $\hat{n}$ of an NLC is perpendicular to the electrodes which same as the $x$-$y$ plane. When the ac voltage $V$ applied to the homeotropic NLC system, the first transition called the Fréedericksz transition occurs at $V \geq V_f$, below the critical threshold for convection $V_C$. The director $\hat{n}$ tilts with respect to the $z$-axis by the transition, and its projection on the $x$-$y$ plane is called the $\vec{C}(r)$-director, where $\vec{r} = (x, y)$. The direction of $\vec{C}(r)$ which behaves as Nambu-Goldstone mode can be freely rotated, because of the same energy [4]. If the ac voltage $V$ applied to the system beyond the point of second transition $V_C$, the electroconvection occurs and exhibits a stripe pattern because of the existences of a homogenous wave vector $\vec{q}(r)$. The
nonlinear interaction between the resulting convective modes $\tilde{q}$ and the Nambu-Goldstone modes $\tilde{C}$ leads to the SMT [5]. To inform how far the system of SMT moves from the critical point ($V_c$), some researchers [4, 6, 7] proposed the normalized voltage $\varepsilon$ as a control parameter, defined by $\varepsilon \equiv (V^2 - V_C^2)/V_C^2$.

There are two types of stripe patterns in the electroconvection of the NLC in the homeotropic system, respectively to the ac frequency $f$ of the applied voltage, namely oblique rolls (OR) and normal rolls (NR). Both patterns are separated by a point of frequency called the Lifshitz frequency ($f_L$). Below the $f_L$ lies OR regime, while beyond $f_L$ is for NR regime. For distinguish both regimes, a line defect called blacklines are usually used. Blacklines found only in the OR regime.

We are interested in investigating the phase transition between the regime of NR and OR by extracting the statistical properties of both patterns relaxation using autocorrelation function. In the SMT, the autocorrelation function shows a decay behavior, and usually, it is approached by a simple exponential decay as a type of the chaotic system [6]. But, recently some studies [7] found that simple exponential equation cannot perfectly fit the autocorrelation function of the SMT relaxation. According to that reason, we also tried to use an alternative fitting form for the whole $\varepsilon$ using the Kohlrausch-William-Watts (KWW) function, a generalization equation of the simple exponential decay by introducing the Kohlrausch exponent ($\beta$).

2. EXPERIMENTAL SETUP

The experimental set up is similar to that in ref. [6, 7]. We carried out our experiments by using the nematic liquid crystal $p$-methoxybenzilidene-$p'$-$n$-butylaniline (MBBA) with doping of tetra-$n$-butyle-ammonium bromide (TBAB). MBBA is filled between two parallel glass plates whose surfaces are coated with transparent electrodes, indium tin oxide (ITO). We used rectangular electrodes with different size; there are 3 x 1.5 cm$^2$ and 2 x 2.5 cm$^2$ which we set up into a sandwiched cell. The space between the electrodes is maintained with polymer spacers of 50 $\mu$m. To realize homeotropic alignment surfaces of the glass plates are treated with a surfactant ($n$-$n$-$dimethyl-n-octadecyl-3-amino-propyl-trimethoxy silyl chloride: DMOAP). Statistical data were measured at the stabilized temperature $30.00 \pm 0.05^\circ$C. The CCD camera (Panasonic BD400) mounted on the microscope, and the software (Pixel View) were used to capture pattern images on the $x$-$y$ plane. Then, the image analysis was performed by custom software.

![Figure 1. The pattern formation at $f = 950$ Hz (a) NR pattern when the control parameter is $\varepsilon = 0.050$ (b) the transition regime between NR and OR is at $\varepsilon = 0.075$, indicated by the appearance of blacklines.](image)

By observing the pattern with the change of frequency and the voltage simultaneously, we obtained $f_L$ for our cell is 750 Hz which separates the OR ($f < f_L$) and the NR regimes ($f > f_L$). We cautiously jumped the voltage until we obtained the transition regime between OR and NR as shown in Figure 1. Moreover, we obtained the phase diagram of the applied ac voltage – frequency ($V$–$f$), respectively. For extracting the statistical properties of the transition NR-OR regime, we set the observation frequency in the Lifshitz frequency $f_L = 750$ Hz. The experimental procedure is as mention in Ref [7]. We set the desired positive control parameter $\varepsilon$ (0.025-0.075) by jumping to the voltage above a convective threshold $V_c$. We captured the electroconvection images in 20 min. Then, we calculated the autocorrelation function $\tilde{Q}(r, \tau)$ of the pattern fluctuation in the Eulerian picture using formula as mentioned in Ref [7].
3. RESULT AND DISCUSSION

The first experimental result of our procedure is the phase diagram for the normal and oblique regime. Figure 2 shows the phase diagram of the applied ac voltage – frequency \((V - f)\), respectively for our cell. The resulting phase diagram from our observation is similar to the schematic phase diagram proposed in the Ref [2]. From Figure 2 we can conclude that the state of NR regime is narrow, proofed by when we set the applied frequency at \(f = 950\ \text{Hz}\) the NR regime appeared below the control parameter \(\varepsilon = 0.075\) and experienced a transition to OR regime at \(\varepsilon = 0.075\) as shown in Figure 2.

![Figure 2](image)

**Figure 2.** Phase diagram of \(V - f\) obtained from observation. The Lifshitz frequency \(f_L\) is about 750 Hz.

While for the experimental results of the temporal autocorrelation analysis for \(0.025 \leq \varepsilon \leq 0.075\) are shown in Figure 3, where \(\tilde{Q}(\tau)\) denotes the spatially average \(\tilde{Q}(r,\tau)\). All the obtained autocorrelation functions fall of rapidly to zero. Also the decay goes faster by increasing of the control parameter \(\varepsilon\), meaning that the dynamical of pattern fluctuation in NR regime is chaotic. Although the autocorrelation function at \(\varepsilon = 0.050\) and 0.075 are coincide. Similar with Ref [7], we also found that the simple exponential is not suitable for describing \(\tilde{Q}(\tau)\) at low \(\varepsilon\). Because of that, we used the KWW equation for fitting function [7] which defined:

\[
\tilde{Q}(r,\tau) = \alpha \exp[-(|r|/\tau_{KWW})^\beta] \tag{1}
\]

where each \(\alpha, \beta,\) and \(\tau_{KWW}\) denotes a normalized constant, the Kohlrausch exponent and the relaxation time of the KWW function. The KWW relation is empirical for relaxation in disorder system and usually used to explain the dynamic in colloidal system. As illustrated in Figure 3, the KWW function well describes the autocorrelation function of SMT for low \(\varepsilon\).

There are two kinds of parameters that we used to describe the effect of the fitting by the KWW relation quantitatively; \(\beta\) and \(\tau_{KWW}\). Figure 4 (a) shows an exponent \(\beta\) dependence with increasing of \(\varepsilon\). If \(\beta > 1\) the KWW equation is called the compressed exponential, while for \(\beta < 1\) is called the stretched exponential. As illustrated in Figure 4 (a), the \(\beta\) value for the transition regime from NR to OR when \(\varepsilon \rightarrow 0\) is greater than 1. Implying that at low \(\varepsilon\) the dynamical of the SMT’s temporal fluctuation is analog with relaxation which appears in glass forming liquid (GFL) dynamics [7]. This dynamical heterogeneity in the GFL has been discussed in Ref. [8] before.

Then, we discuss \(\tau_{KWW}\) parameter or the correlation time proposed by Nugroho et al. [7] as a correction of the correlation time \(\tau_c\) by Hidaka et al. [6] regarding fitting the autocorrelation function to the simple exponential equation. We obtained that the KWW fitting satisfied the relationship \(\tau_{KWW} \propto \varepsilon^{-1}\) as illustrated in Figure 4; namely \(c_0 = -0.00\ \text{s}^{-1}\) defined in \(\tau_{KWW}^{-1} = c_0 + c_1\varepsilon\), showing the bifurcation of the
Also, this was employed as evidence that softening of irregular modes occurs at critical point $\epsilon = 0$. SMT that occurs supercritically and the bifurcation point coincides with electroconvection threshold $\epsilon = 0$.

![Figure 3](image1)

**Figure 3.** The plot of the scaled autocorrelation function with the KWW fitting (shown by a line that intersects with the plotted data) for several values of the control parameters; $\epsilon = 0.025$ (red circle), 0.050 (blue square), and 0.075 (green diamond). Each normalization constant was obtained by fitting. The R value of the fitting is around 0.99, representing perfect fit between the data and the KWW function.

![Figure 4](image2)

**Figure 4.** An $\epsilon$ dependence of $\beta$ (a) and $\tau_{KWW}$ (b) in Eq (2). Standard deviation is used as an error bar.

To get more information about the phase transition from NR to OR we tried to do observation in the NR at higher fixed frequency $f = 1500$ Hz with a convective threshold $V_C$ is 15.9 volt. We expected that we would see another transition from NR to OR. But something different has happened. We didn’t find a sign of transition from NR to OR even we applied a high control parameter $\epsilon = 1.0$ in the system. Furthermore, it is really surprising that we observed a new type of defect appears in the NR regime starts from $\epsilon = 0.8$. There are no studies before that reported a discovery of defects in the NR regime. Defects in the SMT frequently find in the OR regime which called by blacklines. The existence of blacklines is related to the local interaction between $C(r)$ and $q(r)$ that have been discussed before in Ref [5]. Blacklines can be observed for the whole $\epsilon$, and more blacklines will appear by increasing of the control parameter $\epsilon$. It is different with the defects we found in the NR regime. Those defects are very dynamic. In the first appearance the defects take a ring shape, but after several seconds their shape is changed into a point before they begin to disappear. We called those new type of defects by blackspot as shown in Figure 5.
Blackspot only appears when the SMT system is applied by a high control parameter, in random positions and has a short lifetime.

Figure 5. The appearances of the blackspots in the NR regime when $\epsilon = 1.0$, (a) 85 s, (b) 87 s, (c) 90 s, (d) 92 s from initial observation time. The blackspots have a short lifetime.

4. CONCLUSION

We investigated the phase transition in the SMT from NR to OR regime by examined the temporal autocorrelation function of its pattern fluctuation. Our research revealed that in the relaxation of the SMT, there is a GFL-dynamics at low $\epsilon$ indicated by the compressed exponential of KWW which is suitable for fitting the autocorrelation function. Furthermore, at a higher frequency beyond of $f_c$, we found that there is no transition from NR to OR. On the other hand, in the NR regime, we observed new kind of defects called blackspot when the systems applied by a high control parameter. The future researchers are needed to conduct a broader investigation of blackspots properties.

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