Glassy dynamics in relaxation of soft-mode turbulence

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The autocorrelation function of pattern fluctuation is used to study soft-mode turbulence (SMT), a spatiotemporal chaos observed in homeotropic nematics. We show that relaxation near the electroconvection threshold deviates from the exponential. To describe this relaxation, we propose a compressed exponential appearing in dynamics of glass forming liquids. Our findings suggest that coherent motion contributes to SMT dynamics. We also confirmed that characteristic time is inversely proportional to electroconvection’s control parameter.

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Spatiotemporal chaos has been observed and studied in spatially extended nonlinear systems [1]. More than a decade has passed since a spatiotemporal chaos was discovered in homeotropic nematics [2–5]. This type of spatiotemporal chaos was named soft-mode turbulence (SMT) in reference to the softening of the state’s macroscopic fluctuations [4, 5].

SMT is a well-studied nonlinear phenomenon and remains of interest to many researchers. Several studies have focused on the SMT relaxation characterized by the autocorrelation function [4, 9]; in these studies, the simple exponential has been conventionally employed to describe the relaxation. Further, a softening relationship \( \tau_s \propto \varepsilon^{-1} \) has been tested by using the time scale \( \tau_s \) extracted from the simple exponential, where \( \varepsilon \) denotes the control parameter of the electroconvection. However, deviation from the simple exponential has been suggested theoretically [10].

We cautiously investigated the temporal autocorrelation function of SMT’s pattern fluctuation, and show that the relaxation deviates from the simple exponential decay as the system approaches the electroconvection transition point (\( \varepsilon \rightarrow +0 \)). We thus find an alternative fitting form relating to the dynamics of glass forming liquid (GFL) and propose that there exists a similarity between SMT and GFL dynamics. GFL is an example of systems in which relaxation can be nonlinear [11–13]. It is known that dynamical heterogeneity, in which the spatiotemporal fluctuation due to the cooperative effects arises in local transient domains, appears in the vicinity of the glass transition and is a significant concept to understand mechanism of the glass transition [11, 14, 15]. Several works have already pointed out similarity between chaotic and glassy dynamics qualitatively [16–20]; our study experimentally provides quantitative evidences for the similarity. SMT consists of randomly oriented roll domains and shows patch structures [21, 22] which may be strongly related to GFL properties such as the dynamical heterogeneity. In this Letter, we discuss our results from these viewpoints. Moreover, the softening relationship is verified with the time scale defined in the alternative equation.

In the systems in which the director \( \mathbf{n} \) of a nematic liquid crystal is anchored perpendicular to the electrodes, continuous rotational symmetry exists on the plane (\( x-y \) plane) parallel to the electrodes. Applying a magnitude \( V \) of the AC voltage above the Fréedericksz threshold \( V_F \), the director \( \mathbf{n} \) tilts with respect to the \( z \)-axis because the nematic liquid crystal we used has negative dielectric anisotropy, thus breaking the continuous rotational symmetry. The projection \( C(\mathbf{r}) \) of \( \mathbf{n} \) onto the \( x-y \) plane can freely rotate on the \( x-y \) plane because the azimuthal angle of \( C(\mathbf{r}) \) behaves as Nambu–Goldstone mode [2, 23, 24], where \( \mathbf{r} \) denotes the two-dimensional position vector on the \( x-y \) plane. Electroconvection occurs when the applied voltage is above the threshold value \( V_c \), triggering the Carr–Helfrich instability [25]. It interacts with \( C(\mathbf{r}) \) [21], yielding SMT [4, 26]. In this chaotic state, the convective rolls were kept locally and their wave vector \( q(\mathbf{r}) \) was isotropic over the whole system (see figures in Ref. [4]).

The experimental setup is similar to that in Ref. [7], see Fig. 3 therein. We carried out our experiments by using a nematic liquid crystal \( N \)–(4-Methoxybenzylidene)–4-butylaniline (MBBA) sandwiched between two glass plates with the distance \( 52 \pm 1 \) \( \mu \)m. We used circular electrodes, an indium tin oxide (ITO) with diameter 12.9 mm, that is laid on the glass plates. Statistical data were measured at the stabilized temperature 30.00 \( \pm \) 0.05°C. The dielectric constant parallel to the director was \( \varepsilon_{||} = 6.25 \pm 0.1 \), and the electric conductivity parallel to the director was \( \sigma_{||} = (1.17 \pm 0.1) \times 10^{-7} \ \Omega^{-1} \text{m}^{-1} \). The CCD camera (QImaging Retiga 2000R-Sy) mounted on the microscope and the software (QCAPTURE PRO v.5) were used to capture pattern images on the \( x-y \) plane. The resultant image size was 1.14 \( \times \) 1.14 \( \text{mm}^2 \) (i.e., 1000 \( \times \) 1000 pixels).

An AC voltage \( V_{AC}(t) = \sqrt{2}V \cos(2\pi ft) \) was applied to the sample. Two types of SMT pattern arise in response to a specific frequency; namely oblique rolls in \( f < f_l \) and normal rolls in \( f > f_l \) [4, 5], where \( f_l \) denotes the Lifshitz frequency. We set a fixed frequency of \( f = 100 \text{ Hz} \) for SMT pattern to be the oblique roll, where \( f_l = 650 \text{ Hz} \) in our sample cell. The voltage \( V \) was first set between \( V_F = 3.40 \text{ V} \) and \( V_F = 7.82 \text{ V} \), and we subsequently waited 10 min until \( C(\mathbf{r}) \)-director reached a homogenous state [27]. After the homogeneous state was reached, we instantaneously increased \( V \) beyond \( V_c \) in order to obtain the desired value of the normalized control parameter \( \varepsilon = (V/V_c)^2 - 1 \) of the electroconvection, and we then waited 20 min to avoid a transient state.

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$N_T = 600$ images were captured every $\Delta T = 1$ sec. We observed in the above tests that SMT patterns were more disordered and their dynamics quickened with increasing $\varepsilon$, as previously discussed in Ref. [5]. To extract such statistical properties, we calculated the autocorrelation function $\tilde{Q}(r, \tau)$ of the pattern fluctuation in the Eulerian picture;

$$
\tilde{Q}(r, \tau) = \langle (\Delta I(r, t + \tau) - \langle I(r, t) \rangle_t) \Delta I(r, t)^2 \rangle_t^{-1}
$$

with $\Delta I(r, \tau) := I(r, \tau) - \langle I(r, t) \rangle_t$, where $I(r, t)$ denotes the intensity of transmitted light on a position $r$ at time $t$ and the brackets $\langle \rangle_t$ averaging over time. Specifically, each local function $\tilde{Q}(r, \tau)$ at 100 spatial points $r$ was first calculated by averaging over the observation time $T = NT\Delta T$. The obtained set of $\tilde{Q}(r, \tau)$ was then averaged over the 100 spatial points to smooth errors caused by finite observation time. The $10 \times 10$ points were equally spaced in the $x$-$y$ plane.

**Result and Discussion.**—Experimental results of the temporal autocorrelation profiles are shown in Fig. 1 where $\tilde{Q}(\tau)$ denotes the spatially averaged $\tilde{Q}(r, \tau)$. The sudden decrease appears in the short-time regime ($\tau < 1$ sec). It is observed at low $\varepsilon$ and the mechanism has not been elucidated to date. One may need measurements with smaller $\Delta T$ to clarify the specific actions of this mechanism; we do not go into the details of this regime in this Letter. Note that GFL also present such a short-time dynamics [11] [12] and natures of the short-time dynamics do not affect universal behavior of the long-time regime [28] [29].

Temporal autocorrelation functions have been conventionally described in terms of an exponential decay $Q(\tau) \propto \exp[-|\tau|/\tau_c]$. If the dynamics of the pattern fluctuation is Markovian, then the relaxation would be described by the simple exponential. Here, we carefully analyzed the relaxation behavior and found that the simple exponential is not suitable for describing $Q(\tau)$ at a low $\varepsilon$, meaning that SMT dynamics at a low $\varepsilon$ is dominated by the non-Markov process. It is difficult to identify the relaxation form *ab initio*; nevertheless, it may be possible to make predictions to some extent from experimental results. First, one should take into account the fact that SMT relaxations at a high $\varepsilon$ are well-described by the simple exponential. In addition, there is no sign of any transition above the electroconvection transition. Thus, it is expected that the SMT relaxation for whole $\varepsilon > 0$ can be described by an individual function that converges to the simple exponential at a high $\varepsilon$. One possibility for such a fitting function is the Kohlrausch–Williams–Watts (KWW) equation [30] [31]

$$
\tilde{Q}(\tau) = \alpha \exp\left[-(|\tau|/\tau_{\text{KWW}})^\beta\right],
$$

where $\alpha$ denotes a normalized constant, $\beta$ the so-called Kohlrausch exponent, and $\tau_{\text{KWW}}$ the relaxation time of the KWW function. The coefficient $\alpha$ was introduced in Eq. (2) to eliminate the short-time dynamics. The KWW relation is empirical for relaxation in disordered systems (e.g., Ref. [13] as a review). As illustrated in Fig. 1 the KWW relation well describes the SMT dynamics for whole $\varepsilon$ [32].

Figure 2 shows an $\varepsilon$-dependence of the exponent $\beta$. The KWW equation is called the compressed exponential for $\beta > 1$ or the stretched exponential for $\beta < 1$. In the investigated control parameter range, a deviation from simple exponential behavior (i.e., $\beta = 1$) has been observed in a low $\varepsilon$. The deviation seems to increase monotonically with decreasing $\varepsilon$; no jumps nor convergence were found in our results at a low $\varepsilon$. Note that the exponent $\beta \approx 3/2$ has been suggested in a microscopic model [33] [34], where the dynamics is mainly dominated by the random appearance of localized rearrangements; however, no specific features were observed at $\beta \approx 3/2$ in our results. On the other hand, it is reasonable to expect that the exponent $\beta$ converges to unity with increasing $\varepsilon$ because the temporal autocorrelation function can be fitted by the simple exponential at high control parameters, i.e., the stretched exponential is not observed in SMT. This convergence at a high $\varepsilon$ was also confirmed via power-spectrum analysis. It remains unclear to which models the compressed ex-
between correlation time and the control parameter corresponds as evidence that softening of the irregular modes occurs at supercriticality of the SMT bifurcation; this was employed shown in Fig. 2. In dynamics of GFL, the length scale approaches the glass transition point \([42]\). On the other hand, it is supported by the fact that the average patch size increases worthy that the non-thermal fluctuation caused by the patch fluctuation in SMT, as supported by the Lagrangian picture \([21, 41]\). It is consequently noteworthy that the non-thermal noise in SMT is classified as a non-Markov process. That is, the memory effects cannot be negligible. According to the fluctuation-dissipation relation, our result can also be interpreted as showing the emergence of non-thermal noise in SMT, as supported by the Lagrangian picture \([21, 41]\). It is consequently noteworthy that the non-thermal fluctuation caused by the patch structure leads to non-exponential relaxation. This argument is supported by the fact that the average patch size increases with decreasing \(\varepsilon\) \([23]\), which is consistent with the results shown in Fig. 2. In dynamics of GFL, the length scale characterizing dynamical heterogeneity also increases as the system approaches the glass transition point \([42]\). On the other hand, as \(\varepsilon\) increases in SMT, roll structures are easily disturbed and then the lifetime of a roll structure shortens. The ballistic dynamics eventually eliminates at a high \(\varepsilon\), leading to the memory function represented by the delta function; i.e., relaxation can be described by the simple exponential at a high \(\varepsilon\). It is also consistent that dynamical heterogeneity lifetime shortens as the system departs from the glass transition \([43]\).

We next discuss the correlation time \(\tau_{\text{KWW}}\). Kai et al. have experimentally shown that the correlation time \(\tau_s\) diverges as \(\tau_s \propto \varepsilon^{-1}\) \([4]\). It has been discussed that the relationship between correlation time and the control parameter corresponds to supercriticality of the SMT bifurcation; this was employed as evidence that softening of the irregular modes occurs at \(\varepsilon = 0\). However, since we have revealed that the physically meaningful time scale is not \(\tau_s\) but \(\tau_{\text{KWW}}\), we should reexamine whether the softening relation with \(\tau_{\text{KWW}}\) holds. The result obtained from the compressed exponential function satisfies the relationship \(\tau_{\text{KWW}} \propto \varepsilon^{-1}\) shown in Fig. 3; namely, \(c_0 = 0.00 \text{ sec}^{-1}\) and \(c_1 = 0.48 \pm 0.017 \text{ sec}^{-1}\).

![Figure 3](image)

**FIG. 3.** An \(\varepsilon\)-dependence of \(\tau_{\text{KWW}}\) in Eq. (2). The dashed line represents a fitting line: \(\tau_{\text{KWW}}^{-1} = c_0 + c_1 \varepsilon\) with \(c_0 = 0.00 \pm 0.0031 \text{ sec}^{-1}\) and \(c_1 = 0.48 \pm 0.017 \text{ sec}^{-1}\).
law. We thus tried to fit our data by the power law; however, the power law is not suitable for our two-dimensional SMT. To clarify details, one should investigate modal (i.e., wave-number dependent) autocorrelation functions. The temporal autocorrelation function shown in this Letter is regarded as a wave-number average of the modal one \[^52\]. We will discuss SMT’s modal autocorrelation function elsewhere \[^44\]. In addition, spatial dimension might have an effect on the relaxation shape; future research should tackle differences related to the spatial dimension. Further advancement of higher-dimensional models such as those described in Refs. \[^48,50\] is similarly desired.

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