

Universal Fluctuations of Growing Interfaces: Evidence in Turbulent Liquid Crystals

Kazumasa A. Takeuchi* and Masaki Sano

Department of Physics, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

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We investigate growing interfaces of topological-defect turbulence in the electroconvection of nematic liquid crystals. The interfaces exhibit self-affine roughening characterized by both spatial and temporal scaling laws of the Kardar-Parisi-Zhang theory in $1 + 1$ dimensions. Moreover, we reveal that the distribution and the two-point correlation of the interface fluctuations are universal ones governed by the largest eigenvalue of random matrices. This provides quantitative experimental evidence of the universality prescribing detailed information of scale-invariant fluctuations.

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Growth phenomena have been a subject of extensive studies in physics and beyond, because of their ubiquity in nature and their importance in both engineering and fundamental science. Over recent decades, physicists have found that growth phenomena due to local processes typically lead to the formation of rough self-affine interfaces, as exemplified in paper wetting, burning fronts, bacterial colonies, and material morphology, to name but a few, and also in various numerical models [1]. Being obviously irreversible, local growth processes provide a challenging situation toward understanding the scale invariance and the consequent universality out of equilibrium.

The roughness of interfaces is often quantified by their width $w(l, t)$ defined as the standard deviation of the interface height $h(x, t)$ over a length scale l at time t . The self-affinity of interfaces then implies the following Family-Vicsek scaling [2]:

$$w(l, t) \sim t^\beta F(lt^{-1/z}) \sim \begin{cases} l^\alpha & \text{for } l \ll l_* \\ t^\beta & \text{for } l \gg l_* \end{cases} \quad (1)$$

with two characteristic exponents α and β , the dynamic exponent $z \equiv \alpha/\beta$, and a crossover length scale $l_* \sim t^{1/z}$.

The simplest theory to describe such local growth processes was proposed by Kardar, Parisi, and Zhang (KPZ) [3] on the basis of the coarse-grained stochastic equation

$$\frac{\partial}{\partial t} h(x, t) = v_0 + \nu \nabla^2 h + \frac{\lambda}{2} (\nabla h)^2 + \xi(x, t) \quad (3)$$

with $\langle \xi(x, t) \rangle = 0$ and $\langle \xi(x, t) \xi(x', t') \rangle = D \delta(x - x') \delta(t - t')$. For $1 + 1$ dimensions, the renormalization group approach provides exact values of the exponents at $\alpha^{\text{KPZ}} = 1/2$ and $\beta^{\text{KPZ}} = 1/3$ [1,3], which are universal as widely confirmed in numerical models [1]. Moreover, the $(1 + 1)$ -dimensional KPZ class attracts growing interest thanks to rigorous work on the asymptotic form of the fluctuations in solvable models [4–6]. This opens up a new aspect in the study of scale-invariant phenomena toward the universality beyond the scaling laws.

In contrast with such remarkable progress in theory, the situation in experiments has been quite different. A con-

siderable number of experiments have been performed on various growth processes [1] and confirmed the ubiquity of rough interfaces. Concerning the universality, however, experimentally measured values of the exponents are widely diverse and mostly far from the KPZ values for both α and β [1]. To our knowledge, only two experiments among dozens directly found the KPZ exponents: in colonies of mutant bacteria [7] and in slow combustion of paper [8]; a few other experiments showed indirect indications [9,10]. One of the main difficulties shared by most experiments, including the above two, is that one needs to repeat a large number of experiments in the same controlled conditions to accumulate sufficient statistics. In this Letter, studying growing interfaces of turbulent liquid crystals, we overcome this difficulty and report clear experimental evidence of not only the universal scaling laws but also the universal fluctuations of the KPZ class through critical comparisons with the wealth of theoretical predictions.

The electroconvection occurs when an external voltage is applied to a thin layer of nematic liquid crystal, triggering the Carr-Helfrich instability [11]. We focus on interfaces between two topologically different turbulent states called the dynamic scattering modes 1 and 2 (DSM1 and DSM2), which are observed with sufficiently large voltages. The essential difference between them lies in the density of topological defects called the disclinations. Upon applying a voltage, we first observe the DSM1 state with practically no defects in the director field, which lasts until a disclination is finally created owing to the breakdown of surface anchoring [12]. This forms a DSM2 cluster composed of a large quantity of disclinations, which are constantly elongated, split, and transported by fluctuating turbulent flow around. While DSM2 may coexist with DSM1 in a regime of spatiotemporal intermittency [13], for larger voltages we observe growing DSM2 clusters driven by the above-mentioned stochastic local contamination processes.

Our experimental setup consists of a quasi-two-dimensional sample cell, an optical microscope, a thermo-controller, and an ultraviolet pulse laser (see Ref. [13] for

detailed descriptions). The cell is made of two parallel glass plates with transparent electrodes, which are spaced by a polyester film of thickness $12 \mu\text{m}$ enclosing a region of $16 \text{ mm} \times 16 \text{ mm}$ for the convection. We chose here the homeotropic alignment of liquid crystals in order to work with isotropic DSM2 growth, which is realized by coating N , N -dimethyl- N -octadecyl-3-aminopropyltrimethoxysilyl chloride uniformly on the electrodes using a spin coater. The cell is then filled with N -(4-methoxybenzylidene)-4-butylaniline doped with 0.01 wt. % of tetra- n -butylammonium bromide. The cutoff frequency of the conductive regime [11] is $850 \pm 50 \text{ Hz}$. The cell is maintained at a constant temperature 25.0°C with typical fluctuations in the order of 10^{-3} K . The convection is observed through the transmitted light from light-emitting diodes and recorded by a CCD camera.

For each run we apply a voltage of 26 V at 250 Hz, which is sufficiently larger than the DSM1-DSM2 threshold at 20.7 V. After waiting a few seconds, we shoot into the cell two successive laser pulses of wavelength 355 nm and energy 6 nJ to trigger a DSM2 nucleus [13]. Figure 1 displays typical growth of a DSM2 cluster. We repeat it 563 times to characterize the growth process precisely.

We define the local radius $R(x, t)$ along the circle which denotes the statistically averaged shape of the droplets, as sketched in Fig. 1(b). This measures the interfacial width $w(l, t) \equiv \langle \sqrt{[R(x, t) - \langle R \rangle_l]^2} \rangle_l$ and the height-difference correlation function $C(l, t) \equiv \langle [R(x+l, t) - R(x, t)]^2 \rangle$, where $\langle \cdots \rangle_l$ and $\langle \cdots \rangle$ denote the average over a segment of length l and all over the interface and ensembles, respectively. Both $w(l, t)$ and $C(l, t)^{1/2}$ are common quantities for characterizing the roughness, for which the Family-Vicsek scaling [Eq. (1)] is expected.

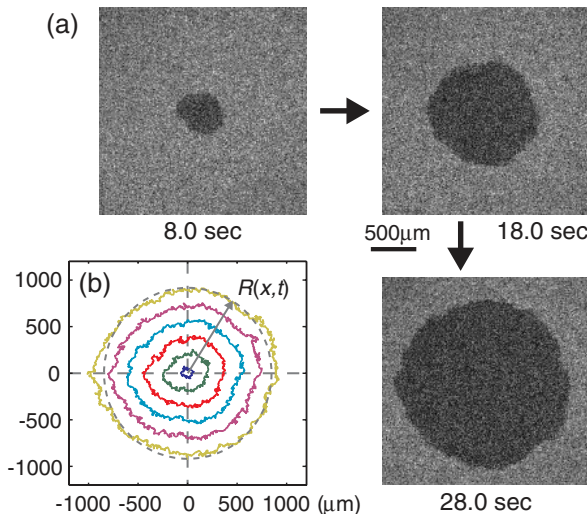


FIG. 1 (color online). Growing DSM2 cluster. (a) Images. Indicated below is the elapsed time after the emission of laser pulses. (b) Snapshots of the interfaces taken every 5 s in the range $2 \text{ s} \leq t \leq 27 \text{ s}$. The gray dashed circle shows the mean radius of all the droplets at $t = 27 \text{ s}$. The coordinate x at this time is defined along this circle.

This is tested in Fig. 2. Raw data of $w(l, t)$ and $C(l, t)^{1/2}$ measured at different times [Figs. 2(a) and 2(b)] grow algebraically for short length scales $l \ll l_*$ and converge to constants for $l \gg l_*$ in agreement with Eq. (1). The power α of the algebraic regime measured in the last frame $t = 28.4 \text{ s}$ is found to be $\alpha = 0.50(5)$. Here, the number in the parentheses indicates the range of error in the last digit, which is estimated both from the uncertainty in a single fit and from the dependence on the fitting range. The found value of α is in good agreement with the KPZ roughness exponent $\alpha^{\text{KPZ}} = 1/2$.

The temporal growth of the roughness is measured by the overall width $W(t) \equiv \sqrt{\langle [R(x, t) - \langle R \rangle]^2 \rangle}$ and the plateau level of the correlation function, $C_{\text{pl}}(t)^{1/2}$, defined as the mean value of $C(l, t)^{1/2}$ in the plateau region of Fig. 2(b). Both quantities show a very clear power law t^β with $\beta = 0.336(11)$ [Fig. 2(c)] in remarkable agreement with the KPZ growth exponent $\beta^{\text{KPZ}} = 1/3$. Furthermore, rescaling both axes in Fig. 2(a) with the KPZ exponents, we confirm that our data of $w(l, t)$ collapse reasonably well onto a single curve [Fig. 2(d)]. A collapse of the same quality is obtained for $C(l, t)^{1/2}$. We therefore safely conclude that the DSM2 interfacial growth belongs to the (1 + 1)-dimensional KPZ class. In passing, this rules out the logarithmic temporal scaling claimed by Escudero for the droplet geometry [14].

Our statistically clean data motivate us to test further predictions on the KPZ class beyond those for the scaling. In this respect one of the most challenging benchmarks may be the asymptotic distribution of height fluctuations, calculated exactly for solvable models [5,6]. A general

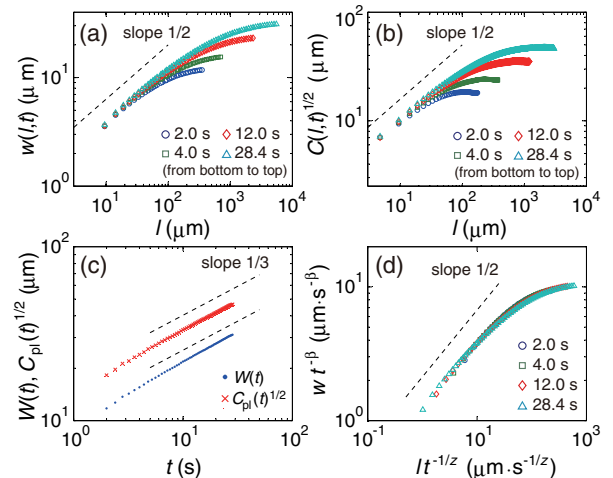


FIG. 2 (color online). Scaling of the width $w(l, t)$ and the height-difference correlation function $C(l, t)$. (a, b) Raw data of $w(l, t)$ (a) and $C(l, t)^{1/2}$ (b) at different times t . The length scale l is varied up to $2\pi\langle R \rangle$ and $\pi\langle R \rangle$, respectively. (c) Time evolution of the overall width $W(t)$ and the plateau level $C_{\text{pl}}(t)^{1/2}$ of the correlation function. (d) Collapse of the data in (a) showing the Family-Vicsek scaling [Eq. (1)]. The dashed lines are guides for the eyes showing the KPZ scaling.

expression was proposed by Prähofer and Spohn [6], which reads $h(t) \simeq v_\infty t + (A^2 \lambda t/2)^{1/3} \chi$ with $A \equiv D/2\nu$, the asymptotic growth rate v_∞ , and a random variable χ obeying the Tracy-Widom (TW) distribution [15], or the (rescaled) largest eigenvalue distribution of large random matrices. The random matrices are from the Gaussian unitary and orthogonal ensemble (GUE and GOE) [16] for curved and flat interfaces, respectively. This implies an intriguing relation to the random matrix theory and requires no fitting parameter provided that the values of the two KPZ parameters λ and A are measured. The prediction was tested once for flat interfaces in the paper combustion experiment [17] with an apparent agreement. However, the authors had to shift and rescale the distribution function for want of the values of the KPZ parameters, in which case the difference among the predicted distributions and the Gaussian one is unpronounced. They also had to discard data subject to intermittent advance of burning fronts due to quenched disorder [17]. Therefore, a quantitative test of Prähofer and Spohn's prediction has not been carried out so far.

We first measure the value of λ experimentally. For the circular interfaces, λ is given as the asymptotic radial growth rate, which has a leading correction term as $\lambda \simeq d\langle R \rangle/dt + a_\nu t^{-2/3}$ for $t \rightarrow \infty$ [18]. This relation is indeed confirmed in Fig. 3(a) and yields a precise estimate at $\lambda = 35.40(23) \mu\text{m/s}$.

The parameter A can be determined, at least for flat interfaces, from the amplitude of $C(l, t)$ and $w(l, t)$ through $C \simeq Al$ and $w^2 \simeq Al/6$ in the limit $t \rightarrow \infty$ [18]. Figure 3(b) shows $C(l, t)/l$ against l for different times t . A similar series of plots is obtained for $6w^2/l$. The value of A can be estimated from the plateau level or the local maximum of these plots, but we find that these estimates increase slowly with time and do not agree with each other (inset). This allows us to have only a rough estimate $A \approx 10 \mu\text{m}$ for the range of time we study.

Now we test Prähofer and Spohn's prediction for the circular interfaces:

$$R(t) \simeq \lambda t + (A^2 \lambda t/2)^{1/3} \chi_{\text{GUE}} \quad (4)$$

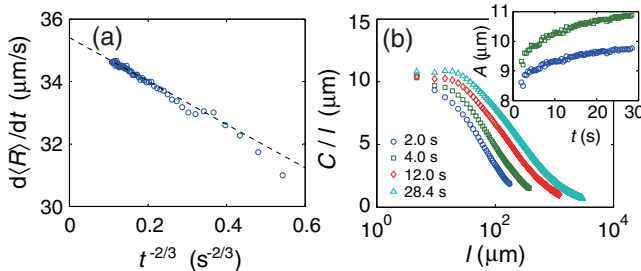


FIG. 3 (color online). Parameter estimation. (a) Growth rate $d\langle R \rangle/dt$ averaged over 1.0 s against $t^{-2/3}$. The y intercept of the linear regression (dashed line) provides an estimate of λ . (b) $C(l, t)/l$ against l for different times t . Inset: nominal estimates of A obtained from $w(l, t)$ (blue bottom symbols) and $C(l, t)$ (green top symbols) as functions of t (see text).

with a random variable χ_{GUE} obeying the GUE TW distribution. We first compute the cumulant $\langle R^n \rangle_c$, for which Eq. (3) implies $\langle R^n \rangle_c \simeq (A^2 \lambda/2)^{n/3} \langle \chi_{\text{GUE}}^n \rangle_c t^{n/3}$ for $n \geq 2$. Our data indeed show this power-law behavior in time [Fig. 4(a)], though higher order cumulants are statistically more demanding and hence provide less conclusive results. We then calculate the skewness $\langle R^3 \rangle_c / \langle R^2 \rangle_c^{3/2}$ and the kurtosis $\langle R^4 \rangle_c / \langle R^2 \rangle_c^2$, which do not depend on the parameter estimates. The result in Fig. 4(b) shows that both amplitude ratios asymptotically converge to the values of the GUE TW distribution, about 0.2241 for the skewness and 0.09345 for the kurtosis [6], and clearly rules out the GOE TW and Gaussian distributions. Conversely, if we admit the GUE TW distribution, the amplitude of $\langle R^2 \rangle_c$ offers a precise estimate of A at $9.98(7) \mu\text{m}$, which is consistent with the direct estimate obtained above and hence used in the following.

Histograms of the local radius $R(x, t)$ are then made and shown in Fig. 4(c) for two different times as functions of $q \equiv (R - \lambda t) / (A^2 \lambda t/2)^{1/3}$, which corresponds to χ_{GUE} if Eq. (3) holds. The experimental distributions show remarkable agreement with the GUE TW one without any fitting, apart from a slight horizontal translation. Indeed, time series of the difference between the n th order cumulants of q and χ_{GUE} [Fig. 4(d)] reveal that the second to fourth order cumulants of q converge quickly to the GUE TW

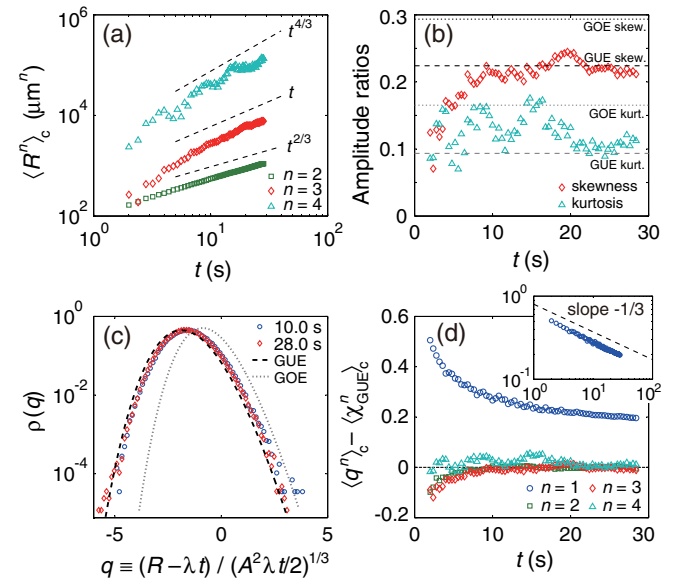


FIG. 4 (color online). Local radius distributions. (a) Cumulants $\langle R^n \rangle_c$ vs t . The dashed lines are guides for the eyes showing the indicated powers. (b) Skewness $\langle R^3 \rangle_c / \langle R^2 \rangle_c^{3/2}$ and kurtosis $\langle R^4 \rangle_c / \langle R^2 \rangle_c^2$. The dashed and dotted lines indicate the values of the skewness and the kurtosis of the GUE and GOE TW distributions. (c) Local radius distributions as functions of $q \equiv (R - \lambda t) / (A^2 \lambda t/2)^{1/3}$. The dashed and dotted lines show the GUE and GOE TW distributions, respectively. (d) Differences in the cumulants of q and χ_{GUE} . The dashed line indicates $\langle q^n \rangle_c = \langle \chi_{\text{GUE}}^n \rangle_c$. Inset: the same data for $n = 1$ in logarithmic scales. The dashed line is a guide for the eyes.

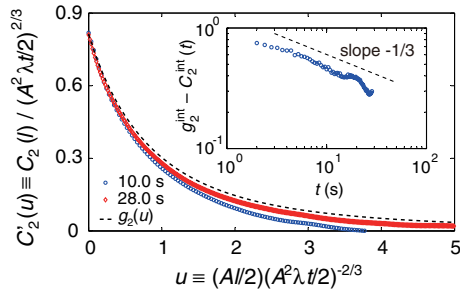


FIG. 5 (color online). Two-point correlation function $C_2(l, t)$ plotted in the rescaled axes $u \equiv (Al/2)(A^2\lambda t/2)^{-2/3}$ and $C_2'(u) \equiv C_2(l)/(A^2\lambda t/2)^{2/3}$. The dashed line indicates the theoretical prediction $g_2(u)$. Inset: integral of the rescaled correlation function $C_2^{\text{int}} \equiv \int_0^\infty C_2'(u)du$ as a function of time t . The difference from the theoretical counterpart $g_2^{\text{int}} \equiv \int_0^\infty g_2(u)du$ is shown. The dashed line is a guide for the eyes.

values, while the first order one, i.e., the mean, algebraically approaches it with a power close to $-1/3$ (inset). This is theoretically reasonable behavior which stems from the existence of an additional constant term in Eq. (3). Therefore, we conclude that the local radii of the DSM2 nuclei asymptotically obey the GUE TW distribution at least up to the fourth order cumulants, confirming the prediction of Prähofer and Spohn.

We also measure the two-point correlation function $C_2(l, t) \equiv \langle R(x+l, t)R(x, t) \rangle - \langle R \rangle^2$. Theory predicts that $C_2(l, t)$ is asymptotically described by the $\mathcal{A}_2(t)$ process or by the dynamics of the largest eigenvalue in Dyson's Brownian motion of GUE matrices [16] as $C_2(l, t) \simeq (A^2\lambda t/2)^{2/3} g_2(u)$ with $g_2(u) \equiv \langle \mathcal{A}_2(u+t) \mathcal{A}_2(t) \rangle$ and $u \equiv (Al/2)(A^2\lambda t/2)^{-2/3}$ [19]. Our experimental data confirm this with an algebraic finite-time correction consistent with the power $-1/3$ (Fig. 5).

In comparison with past experimental studies showing diverse scalings, one may wonder why the liquid crystal turbulence exhibits such clear KPZ-class behavior. We consider that the following three factors are essential. (i) The growth of DSM2 results from strictly local processes due to the turbulent flow on the interfaces and not from inward or outward interactions of the cluster, which could induce long-range effects and affect the universality. (ii) The stochasticity of the process stems from intrinsic turbulent fluctuations overwhelming quenched disorder. (iii) Good controllability and fast response of the liquid crystals allowed us to repeat hundreds of experiments in the same conditions, leading to statistically reliable data. The reproducibility of the presented results was confirmed with different voltages and spatial resolutions with the same quality of data (not shown).

In conclusion, measuring the growth of DSM2 nuclei in the electroconvection, we have found the circular interface roughening clearly characterized by the scaling laws of the KPZ class in $1+1$ dimensions. Moreover, we have shown without fitting that the fluctuations of the cluster local radius asymptotically obey the Tracy-Widom distribution

of the GUE random matrices and revealed the finite-time effect. Together with the agreement in the two-point correlation, our experimental results quantitatively confirm the geometry-dependent universality of the $(1+1)$ -dimensional KPZ class prescribing detailed information of the scale-invariant fluctuations. In this respect, investigations of flat interfaces in the same system are of outstanding importance and are in progress.

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Note added in proof.—After submission of this Letter, Sasamoto and Spohn reported an exact solution of the $(1+1)$ -dimensional KPZ equation [20], which offers a clear theoretical ground of our experimental results.

*kazumasa@daisy.phys.s.u-tokyo.ac.jp

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