Dynamics of cholesteric liquid crystals in the presence of random magnetic fields

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Abstract – Based on dynamic renormalization group techniques, this letter analyzes the effects of external stochastic perturbations on the dynamical properties of cholesteric liquid crystals, studied in the presence of a random magnetic field. Our analysis quantifies the nature of the temperature dependence of the dynamics; the results also highlight a hitherto unexplored regime in cholesteric liquid-crystal dynamics. We show that stochastic fluctuations drive the system to a second-ordered Kosterlitz-Thouless phase transition point, eventually leading to a Kardar-Parisi-Zhang (KPZ) universality class. The results go beyond quasi–first-order mean-field theories, and provide the first theoretical understanding of a KPZ phase in distorted nematic liquid-crystal dynamics.

A liquid crystal is a special state of matter that shares properties both from a liquid as well as from a (solid) crystal. They are differentiated based on their optical properties, that, in turn, depend both on positional ordering as well as on orientational ordering. In effect, a liquid-crystal sample exhibits a competition between the alignment of the director \(\mathbf{n}\) at a surface or boundary and the orientation induced within the sample by an externally applied electric or magnetic field which produces a spatial rearrangement of the director subjective of the phase concerned [1–3]. A cholesteric (Ch) mesophase is locally very similar to a nematic phase except that it is composed of optically active molecules. When an optically active material forms a nematic phase, the preferred direction of the long-molecular axis varies in a direction throughout the medium in a regular way and displays a continuous twist along the optical axis leading to a helical structure. Thus, the cholesteric phase is a distorted nematic phase. The present article provides the first theoretical model addressing the phase transition properties of a cholesteric liquid crystal perturbed by a stochastic magnetic field.

As noted before, a liquid crystal is characterized by the configurational deformation driven by a magnetic field. In thermodynamic parlance, this is defined as a Freedericksz transition that occurs in a liquid-crystal slab when the director reorientates in the direction of an applied magnetic field larger than a critical field \(H_C\). It is also well known that the Freedericksz instabilities occur in nematic liquid crystals [1–6]. Planar cholesteric films can also exhibit either a Freedericksz transition or a transition to a stripe texture in electric fields [7]. Similar Freedericksz instabilities are expected in cholesteric liquid crystals. Valkov \textit{et al.} [8] studied the Freedericksz transition in the twist cells of cholesteric liquid crystals with a finite surface energy. It was shown that this transition can be either of the second order or of the first order depending on the values of the Frank constants, pitch, surface energies, and the cell thickness [8]. Zumer \textit{et al.} [9] theoretically analyzed the field-induced structural (Freedericksz) transitions in supramicron nematic and cholesteric liquid-crystal droplets embedded in a solid matrix. They obtained the equilibrium structures and phase diagrams by taking into account the elastic, surface, and field free-energy contributions. Experimental work on the dynamics of the Freedericksz transition [10–12] shows the existence of transient spatial structures. Sagues and Miguel [13] studied the Freedericksz transition under twist deformation in a nematic layer when the magnetic field has a random

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component. It was found that the randomness of the field produces a shift in the instability point. Beyond this instability point, the time constant characteristic of the approach to the stationary stable state decreases because of the field fluctuations.

In a remarkable departure from deterministic analysis, Miguel [14] studied the stationary spatial correlations of the director field of a nematic liquid crystal near the Freedericksz transition in a magnetic field with a time-dependent random component. They showed that the correlation length diverges at a shifted instability point. Sagues and Miguel [15] studied transient dynamics of spatial fluctuations of the director field in the pure twist Freedericksz transition. They analyzed linear theories at different stages of evolution and the domain of validity. In a later work [16], the same authors studied the dynamics of a transient pattern formation in the Freedericksz transition corresponding to a twist geometry. They calculated the time-dependent structure factor based on a dynamical model which incorporates consistently the coupling of the director field with the velocity flow and also the effect of fluctuations. They identified a time scale associated with the emergence of the pattern and a subsequent stage of pattern development.

Apart from rare theoretical attempts routed through renormalization group applications [17], none of these aforementioned studies, however, analyze dynamic perturbations of cholesteric liquid crystals in the presence of random magnetic field, as is often seen or expected in experiments [10–12]. The purpose of the present paper is to discuss the dynamical evolution patterns of cholesteric liquid crystals in a random magnetic field using the language of dynamic renormalization groups. We elucidate the main consequences of a fluctuating environment in such dynamical aspects. We show that in the experimentally viable low-frequency limit, that is at large spatiotemporal scales, the dynamical model predicts the onset of a Kosterlitz-Thouless transition from the emergence of an equivalent sine-Gordon model, in agreement with results mapped from different phenomenological models [18,19]. At large enough spatiotemporal scales, the inherent nonlinearity drives the system to a Kardar-Parisi-Zhang fixed point, represented by appropriate self-affine scaling.

We start with a Frank free energy [20], consistent with the phenomenological continuum theory of liquid crystals that is very successful in explaining various magnetic- or electric-field-induced effects. When the external field is applied perpendicularly to the helical axis, at low temperatures, it unwinds the helical structure of the cholesteric state via a nucleation-type transition [1]. The elastic free energy of a cholesteric liquid crystal in the presence of magnetic field can be written as [5]

\[
g_{ch} = \frac{1}{2}K_1(\nabla \cdot \mathbf{n})^2 + \frac{1}{2}K_2(n \cdot (\nabla \times \mathbf{n}) + q_0)^2 + \frac{1}{2}K_3(n \times (\nabla \times \mathbf{n}))^2 - \frac{1}{2}\chi_a(n \cdot \mathbf{H})^2, \tag{1}
\]

where \(K_i (i = 1, 2, 3)\) are three elastic constants associated with splay, twist and bend deformations, respectively. \(\chi_a\) is the anisotropic part of the magnetic susceptibility and \(q_0\) is the wave vector corresponding to the magnetic-field amplitude \(H = 0\) (\(q_0 = 2\pi/\lambda_h\)). \(\lambda_h\) being the pitch of the helix. In the rest of this article, we will always consider positive anisotropy magnetic susceptibility.

Let us start with a cholesteric sample of thickness \(d = \lambda_h/2\). We assume that, at the walls strong anchoring prevails along the \(x\)-axis, which then becomes the direction of the applied external magnetic field \(\mathbf{H}\). The helical axis \(z\) is chosen normal to \(\mathbf{H}\). This defines “pure twist” with the following components of the director \(\mathbf{n}\) orientation: \(n_x = \cos \phi(z,t), n_y = \sin \phi(z,t)\). \(n_z = 0\). \(\phi(z,t)\) is the angle between \(\mathbf{n}\) and the \(x\)-axis. Boundary conditions are \(\phi(z = 0) = 0\) and \(\phi(z = d) = \pi\). Then the non-holonomic elastic free energy \(G\) is given by

\[
G = \frac{S}{2} \int_0^d \left[ \frac{K_2}{2} \left( \frac{d\phi}{dz} - q_0 \right)^2 - \frac{\chi_a H^2}{2} \cos^2 \phi \right] dz = \\
\int L(z, \phi(z), \phi'(z)) dz, \tag{2}
\]

where \(S\) is the surface area of the plates and \(L(z, \phi(z), \phi'(z))\) is the equivalent Lagrangian of the system.

The time-dependent Ginzburg-Landau (TDGL)-type model can be specified by the following Langevin dynamics:

\[
M \frac{\partial \phi(z,t)}{\partial t} = -\frac{\delta G}{\delta \phi} + F_0 + \eta(z,t), \tag{3}
\]

where \(\eta(z,t)\) is a stochastic white noise represented by \(\langle \eta(z,t) \eta(z',t') \rangle = D_0 \delta(z - z')\delta(t - t')\) and \(\langle \eta(z,t) \rangle = 0\), in which the curly brackets \(\langle \cdot \rangle\) represent the ensemble average over all noise realizations and \(D_0\) is the noise strength, a constant, while \(\frac{\delta G}{\delta \phi}\) is the functional derivative of \(G\) with respect to the variable \(\phi\). \(F_0\) is a constant “overdamped force” that pumps a steady energy into the system. While admittedly the original Frank-Oseen model (Frank energy) does not have such a constant force \(F_0\) in its definition, such a term is related to the existence of a finite non-trivial kinetic roughening in the model that can only be counter-balanced by a negative damping term \(-F_0\). This will be later analyzed in detail through a renormalization group analysis.

Equation (3) can then be rewritten as

\[
M \frac{\partial \phi}{\partial t} = \frac{S}{2} \left( -\frac{\chi_a H^2}{2} \sin 2\phi + K_2 \nabla^2 \phi \right) + F_0 + \eta(z,t). \tag{4}
\]

The above equation (4) represents a quintessential sine-Gordon model [21] whose low-frequency spatiotemporal scaling properties have been studied by Chattopadhyay [18]. In what follows, we will consistent use \(\nabla^2 \phi = \frac{\partial^2 \phi}{\partial z^2}\) in order to restrict our analysis to the axial function.
Along the z-direction, the steady-state dynamics of eq. (4) leads to
\[ K_s \frac{d^2 \phi}{dz^2} = \frac{\chi_s H^2}{2} \sin 2\phi - \frac{2F_0}{S} \]
which, for \( F_0 = 0 \), can be easily solved in terms of Jacobi elliptic functions. For a general non-zero \( F_0 \), eq. (5) may be numerically solved.

Equation (4) can be further rescaled, which, on a periodic lattice, leads to the following model:
\[ \eta \frac{\partial h}{\partial t} = \gamma \frac{d^2 h}{dz^2} - \frac{2\pi V}{a} \sin \left( \frac{2\pi}{a} h \right) + F + N(z, t), \]
where \( \eta = \frac{M_\pi \gamma}{k_s H^2} \), \( \gamma = \frac{\pi}{2a} F \), \( F = \frac{F_0}{\eta} \), and the rescaled noise \( N(z, t) = \eta(z, t) / K_s \). The above is the classic noisy sine-Gordon model that can be studied using the method previously used by Chattopadhyay [18].

The model presented in eq. (6) is the classical noisy sine-Gordon model that can be studied in the limit of zero noise perturbations. From eqs. (12a)–(18b), we can now write
\[ \delta h^{(0)}(z, t) = \int_{-\infty}^{t} dt' \int dz'(G_0(z - z', t - t') \delta N(z', t')) \]
where \( u(1-x) = \theta(x) \), a Heaviside step function, \( A \propto \frac{\xi}{a} \), and \( \widetilde{D} = \eta T \). is the Green’s function.
Then it is easy to calculate the first-order renormalization group (RG) flows for $V$ and $F$. In order to calculate the RG flows for $\gamma$, $\eta$ and $D$, we need to evaluate the second-ordered corrections terms $\langle \delta h^{(0)}(z, t) \delta h^{(1)}(z, t) \rangle$ and $\langle \partial_t \delta h^{(0)}(z, t) \partial_t \delta h^{(1)}(z, t) \rangle$. These two terms can be expressed as

$$
\langle \delta h^{(0)}(z, t) \delta h^{(1)}(z, t) \rangle =
-\frac{4\pi^2}{a^2} V \int_{-\infty}^{t} dt' \int dz' \cos \left[ \frac{2\pi}{a} \left( \frac{\hat{h}(z', t') + F'}{\eta} \right) \right] G_0(z - z', t - t')
\times \langle \delta h^{(0)}(z', t') \delta h^{(0)}(z, t) \rangle
\times \left( \frac{z' - z}{t - t'} \right)^{2} G_0(z - z', t - t')
\times \langle \partial_t \delta h^{(0)}(z, t) \partial_t \delta h^{(0)}(z', t') \rangle
\tag{17a}
$$

(17b)

Combining the variable rescaling $t \rightarrow t' = t/\eta$ and $h \rightarrow h' = h - \frac{t}{\eta} \gamma$, with the Rost-Spohn approximation scheme, the second-ordered renormalized $\langle \delta h^{(0)}(z, t) \delta h^{(1)}(z, t) \rangle$ gives

$$
\Psi_{SG} \approx -\frac{8\pi^3 V^2 T}{\gamma^2 a^5} \int_{-\infty}^{t} dt' \int dz' e^{i(\lambda|z - z'|)}
\times e^{-\frac{1}{4\xi} \left( \frac{z' - z}{t - t'} \right)^4}
\frac{2\pi}{a} \left( \frac{\partial}{\partial t} \hat{h}(z, t)(t - t') - \frac{1}{2} \partial_t \partial_t \hat{h}(z, t)(z_i - z_i')(z_j - z_j') \right)
\cos \left( \frac{2\pi F}{a} (t - t') \right)
\sin \left( \frac{2\pi F}{a} (t - t') \right).
\tag{18}
$$

The terms in eq. (18) that are respectively proportional to $\frac{\partial^2}{\partial t^2}$, $\partial_t \partial_t$ and $(\partial_t \hat{h})^2$ are the renormalized contributions for $\eta$, $\gamma$ and a new KPZ nonlinearity $\lambda$ that automatically gets created from a sine-Gordon potential. The constant term above renormalizes $F$.

This leads to the RG flow equations as follows:

$$
\frac{dU}{dt} = (2 - n)U, \tag{19a}
\frac{d\gamma}{dt} = \frac{8\pi^4}{\gamma a^2} n A^{(1)}(n; k) U^2, \tag{19b}
\frac{d\eta}{dt} = \frac{32\pi^4}{\gamma a^2} n A^{(0)}(n; k) U^2, \tag{19c}
\frac{d\lambda}{dt} = \frac{32\pi^5}{\gamma a^5} n A^{(0)}(n; k) U^2, \tag{19d}
\frac{dD}{dt} = \frac{32\pi^4}{\gamma a^4} n A^{(n)}(n; k) U^2, \tag{19e}
\frac{dK}{dt} = 2K - \frac{8\pi^3}{\gamma a^3} n A^{(K)}(n; k) U^2, \tag{19f}
$$

where $U = V/A^2$, $K = F/A^2$, $\bar{\rho} = \Lambda \rho$, $x = \frac{x(t - t')}{\eta}$, $n = \frac{\eta T}{\pi T}$ and $\kappa = \frac{2\pi K}{\eta T}$.

The functional forms of the renormalization group (RG) flow integrals $A^{(\gamma)}(n; k)$, $A^{(\eta)}(n; k)$, $A^{(\lambda)}(n; k)$ and $A^{(K)}(n; k)$ are defined as follows:

$$
A^{(\gamma)}(n; k) = \int_{0}^{\infty} \frac{dx}{x} \int_{0}^{\infty} d\bar{\rho} \bar{\rho}^2 \cos(\bar{\rho}) \cos \left( \frac{2\pi x \bar{\rho}^2}{\gamma} \right) e^{-\bar{\rho}},
$$

$$
A^{(\eta)}(n; k) = \frac{x}{\bar{\rho}^2} A^{(\gamma)}(n; k),
$$

$$
A^{(\lambda)}(n; k) = \int_{0}^{\infty} \frac{dx}{x} \int_{0}^{\infty} d\bar{\rho} \bar{\rho}^2 \cos(\bar{\rho}) \sin \left( \frac{2\pi x \bar{\rho}^2}{\gamma} \right) e^{-\bar{\rho}},
$$

$$
A^{(K)}(n; k) = \frac{1}{\bar{\rho}^2} A^{(\lambda)}(n; k),
$$

$$
f = \frac{1}{2x} + x \bar{\rho}^2 + \frac{2\pi \eta}{\gamma a^2} \chi(\bar{\rho}, x),
$$

$$
\chi(|z - z'|, |t - t'|) = \frac{\pi}{\eta T} \left( \langle \hat{h}(z, t) - \hat{h}(z', t') \rangle \right)^2,
$$

$$
\langle \hat{h}(z, t) - \hat{h}(z', t') \rangle^2 = \eta T \int_{0}^{1} dk \left[ \sqrt{2(1 - \cos k|z - z'|)} \right] e^{-\frac{\pi k^2}{4}(t - t')}.
$$
We now discuss the physical realization of the RG flow equations. The variation of the flow patterns at the fixed point $n = \frac{\pi T}{\eta} = 2$ are depicted in fig. 1; this clearly suggests that as the external force $K$ (or proportionately $F$) increases, both the renormalized diffusion constant $\gamma$ and the forcing strength increase. Since $\gamma \sim \frac{1}{\sqrt{T}}$, as $\gamma$ increases, the non-equilibrium temperature of the system decreases and vice versa. The most interesting aspect of these flows, though, is the emergence of a hitherto unseen Kardar-Parisi-Zhang (KPZ) [22] nonlinearity that steadily increases, the non-equilibrium temperature of the system thereby converging to a Kardar-Parisi-Zhang universality class. The nature of the temperature dependence of the cholesteric phase has also been explained. The model additionally provides the first theoretical confirmation of the recently observed KPZ phase in nematic liquid crystals [23]. Our results are expected to encourage further experiments, including the study of the dynamical properties of other liquid-crystalline phases, focusing now on finite-sized effects of samples, an aspect that we are presently working on.

In complementarity, the friction term $\eta$ gets compressed in the flow space resulting in a decay curve as shown in fig. 1(b). Although we do not have any $(\nabla \phi)^2$ term (KPZ term) in the original dynamics as shown in eq. (4), such a term is generated due to the presence of the sine-Gordon potential in the dynamics. Even if $\lambda = 0$ at the start, the $\frac{d}{dt}$ in eq. (19d) ensures that a nonzero $\lambda$ is eventually generated. Once $\lambda \neq 0$, the effective temperature of the system increases due to renormalization. When $n = \frac{\pi T}{\eta} > 2$, $U$ decays to zero. Thus, at a large enough spatiotemporal scale, the dynamics is governed by KPZ scaling.

The prediction of the KPZ universality class for $1 + 1$ dimensions validates a recent experimental work by Takeuchi and Sano [23] where they studied the scale-invariant fluctuations of growing interfaces in nematic liquid-crystal turbulence. As a perturbed nematic phase, a cholesteric phase too is expected to behave identically.

The flow equations given by eqs. (19a)–(19f) are not valid at very small $T$’s. Although the original model (Frank energy) does not have a constant force ($F$) term, such a term will be generated by a renormalization group analysis of the model. In order to control kinetic roughening, of the cholesteric liquid crystal, especially in the high-temperature regime, an opposing force $-F$ is ingrafted into the dynamics at all times. The results from this analysis, thus identify how to control the dynamics of a cholesteric liquid crystal and what particular combination of parameters is capable of achieving this. Also the results go beyond the usual confines of mean-field analysis, now including the second order RG correction in analyzing the contribution from the dynamical modes.

In conclusion, we have studied the impact of stochastic fluctuations in modifying the spatiotemporal properties of cholesteric liquid crystals in the presence of a random magnetic field. In the mode considered, we focus on strong anchoring of the liquid-crystal molecules to the boundaries. For general studies on surface anchoring, a Rapini-Papoular potential may be considered in line with [24]. The core renormalizability aspect of the theory will still remain intact though and would be perfectly applicable with such potentials, too. Our results show that stochastic fluctuations drive the system to a Kosterlitz-Thouless transition point, through a second-ordered phase transition, thereby converging to a Kardar-Parisi-Zhang universality class.