

Evidence of topological charge polarization at active-passive interfaces in acoustically powered active liquid crystals

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Theoretical and computational studies predict topological charge separation at active-passive interfaces in active nematics, but reliable experimental validation has been lacking. We utilize a synthetic acoustically energized liquid crystal to experimentally investigate the dynamics of topological defects across an active-passive interface. The interference pattern induced by the acoustic wave inside the experimental cell creates a spatial distribution of activity, resulting in the formation of active-passive interfaces with the liquid-crystalline director field aligned parallel to those interfaces. The activity gradient drives the reorientation of positively charged topological defects along its direction, causing them to migrate into the passive zone and form a layer with net topological charge. That layer “discharges” upon cessation of activity through annihilation of positive and negative topological charges, resembling the discharge of a capacitor.

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I. INTRODUCTION

One of the most intriguing and extensively studied phenomena observed in active nematic materials is the continuous proliferation, annihilation, motion, and alignment of topological defects [1–4]—localized singularities of the order parameter where the director field cannot be defined [5]. Defect generation and annihilation occur in pairs of opposite topological charges, $+1/2$ for “cometlike” defects and $-1/2$ for “trianglelike” [6]. The topological defects can be regarded as active entities with the dynamics governed by the local active and passive stresses [2,7–9]. In a passive (equilibrium) liquid crystal, topological defects of opposite sign experience attractive interaction [7], approach each other, and annihilate to minimize the free energy. In active nematic systems, the entire concept of equilibrium thermodynamics is not applicable, and dynamics of topological defects becomes more complex [10–12]. In addition to spontaneous generation and annihilation, $+1/2$ topological defects become motile and often demonstrate ordering [2,12–15].

The behavior of topological defects in nonhomogeneous active materials with modulated activity or coexisting phases (nematic/isotropic) is nontrivial. It was predicted that in the mixtures of active and passive nematics, the nematic-isotropic interfaces become topologically charged [8,16–21]. Computational studies revealed that the nonuniform activity distribution results in defect trapping [18] in the passive zone and the formation of topologically charged layers

[8,16,22,23]. Positive defects, which are significantly more motile than negative defects, align with the activity gradient [19] and move toward the passive area. Yet, the reliable experimental validation of the above predictions has been lacking. Up to very recently, the experimental active nematics suitable for spatial activity modulation were limited to biologically based systems [22]. There, the concept of spatially structured activity was realized by the use of actin filaments and light-sensitive myosin motors. Spatial activity control was leveraged to direct defect dynamics and manipulate long-range flows in an active nematic liquid crystal [22]. The slow response time (~ 1 min), slow dynamics ($\sim 2 \mu\text{m/s}$) combined with the biological aging of the light-activated protein system (occurring within 1 h), and a low defect count within the experimental field of view hindered the reliable resolution of topological defect separation at active-passive interfaces. Recently reported synthetic active liquid crystals powered by acoustic fields [24] overcome all the above deficiencies of the biological active nematics and present a promising platform for the investigation of the topological defect dynamics at active-passive interfaces. In these systems, spatial profile of activity can be engineered through the interference patterns of the applied acoustic field. In addition, the activity there is conveniently controlled by the amplitude of the acoustic field, which can be rapidly adjusted over a wide range, enabling precise control and manipulation of the density and dynamics of topological defects [24].

In this article, we present a detailed experimental investigation of the topological defect dynamics at active-passive interfaces in active liquid crystals. The interference of the standing acoustic wave creates spatial nonuniformity of activity and, therefore, active-passive interfaces with the gradients of activity controlled by the frequency of the excitation signal. The conversion of acoustic energy into local extensile stresses of the liquid-crystalline medium results in the emergence of an undulation instability (with the characteristic wavelengths

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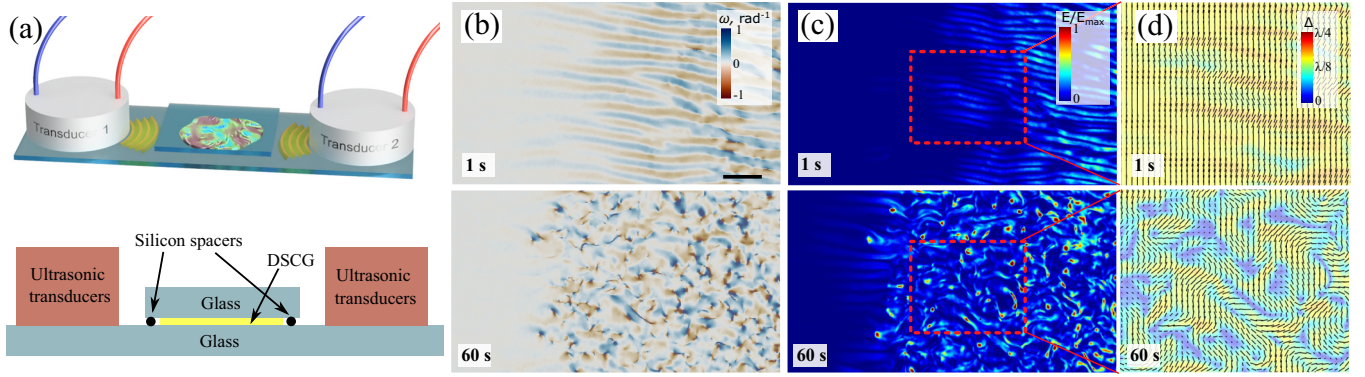


FIG. 1. Formation of active-passive interfaces in an acoustically energized liquid crystal. (a) Schematic representation of the experimental system. The liquid crystal (LC) drop is sandwiched between two glass slides with two ultrasonic transducers attached to the bottom glass. The spatial distribution of the rotation rate of the director field [panel (b)], and (c) elastic energy over the active-passive interface. The order parameter and the director field for the area highlighted in panel (c) are shown in panel (d). The distributions are shown for 1 s (top row) and 60 s (bottom row) after the acoustic field is applied. The scale bar is 50 μm .

governed by the amplitude of the acoustic field) and proliferation of topological defects [24]. Our experiments reveal that positive topological defects migrate across the interface, resulting in a topological charge polarization at the interface. The polarization layer vanishes when the activity is switched off through the annihilation of positive and negative topological charges. These results provide direct experimental validation of theoretical predictions related to defect alignment and charge separation at active nematic materials with a spatial profile of activity.

II. EMERGENCE OF ACTIVITY INTERFACES

The experimental system consists of the lyotropic chromonic liquid crystals DSCG (disodium cromoglycate), sandwiched between two glass plates (6 μm sample thickness) with a pair of ultrasonic transducers attached to the bottom glass of the cell [see Fig. 1(a)]. The system can be driven across a broad frequency range from 20 to 500 kHz. Our experiments on an acoustically energized liquid crystal (AELC) reveal that activity is unevenly distributed across the experimental cell, typically appearing in domains of varying shapes. The size of domains scales inversely with the excitation frequency f . This nonuniformity of activity arises from the interference patterns of the acoustic field, which are influenced by the geometry of the cell and shape of the droplet sample. The activity pattern can also be modulated by adjusting the phase difference between the signals driving the ultrasonic transducers.

To quantify the activity in the system, we utilize the temporal evolution and topology of the director field reconstructed via multipolarized microscopy [25]. In particular, the activity can be inferred from the wavelength of undulation instability [4,6,26,27] in the nematic material, defect density [2], defect generation rate and hydrodynamic vorticity [8,14], or spectral characteristics of elastic energy [28,29]. In our previous study [24], we introduced an alternative approach to measuring local activity by time averaging the magnitude of the director rotational rate $\Omega = \frac{1}{\Delta t S} \int_0^{\Delta t} \int_S |\omega| dS dt$. Here, ω is the local rotational rate of the director field, shown in Fig. 1(b) and Movie S1 in the Supplemental Material [30], S represents the

area of averaging (to decrease noise) with a characteristic size of undulation instability ($\sim 20\text{--}50\ \mu\text{m}$), and Δt denotes the time interval of averaging. In regions with spatially varying activity, this method appears to be the most appropriate as it allows to correctly estimate activity in passive regions with topological defects that were pushed or diffused into the region. For instance, undulation instability becomes a spatially distributed phenomenon with a varying wavelength defined by the local activity [see Figs. 1(c) and 1(d) and Movie S2 in the Supplemental Material [30]]. Consequently, measuring activity using a characteristics defined over a large area becomes problematic. For the data presented in this paper, S was selected as a square $50\ \mu\text{m} \times 50\ \mu\text{m}$, with $\Delta t = 10\ \text{s}$. The results presented below are not sensitive to small variations of these parameters. We estimated the spatial distribution of

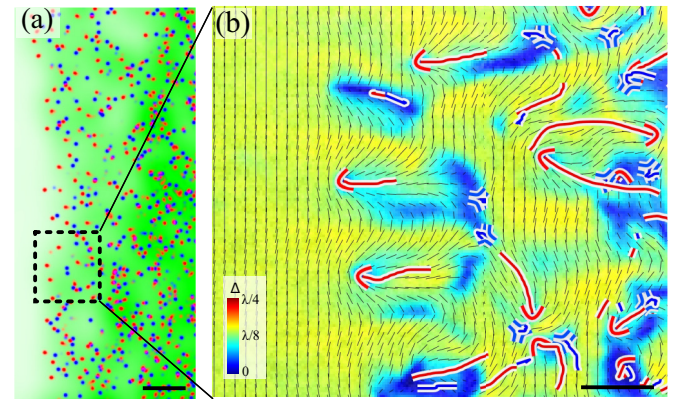


FIG. 2. Topological defects at the active-passive interface. (a) An image of activity distribution over the active-passive interface. The saturation of the green color illustrates the local level of activity in LC. Red and blue spots show positions of $+1/2$ and $-1/2$ topological defects, respectively. Faded spots show previous positions of defects, with the brightness fading rate of $0.1\ \text{s}^{-1}$ (brightness decreased by 90% each second). The scale bar is 200 μm . (b) The local director field orientation, order parameter, and topological defect motion for the area marked by the dashed line in panel (a). The trajectories of defects shown for the last 5 s. The scale bar is 25 μm .

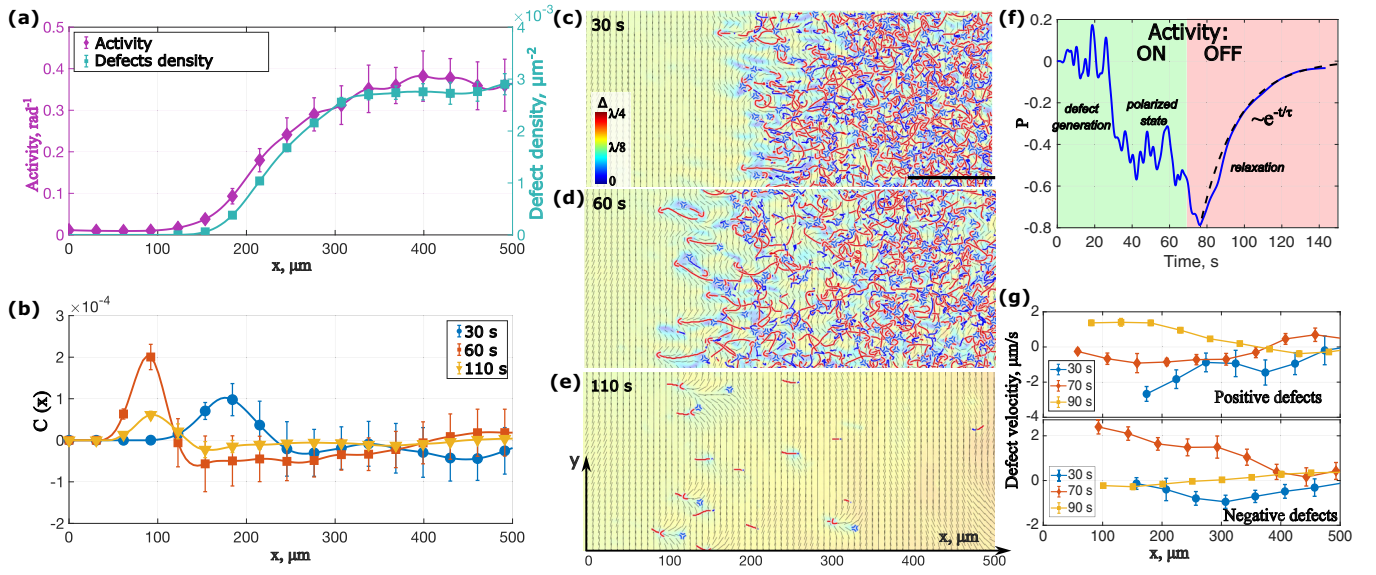


FIG. 3. Topological charge distribution over the activity gradient. For all panels, the activity is turned on at 0 s and turned off at 68 s. The excitation frequency is 61 kHz. (a) Spatial distribution of activity (magenta, diamonds) and the density of all (positive and negative) topological defects (cyan, squares) at 20 s. Gaussian smoothing with the width of 30 μm is applied for the activity profile. (b) The topological charge distribution at different moments of time. Statistics are derived from images $800 \mu\text{m} \times 1000 \mu\text{m}$ averaged over the y direction and 2 s time interval. Adaptive Gaussian smoothing with the width from 5 to 60 μm is applied over x to remove noise (see the Supplemental Material [30] for details). Error bars are standard deviations obtained from the data before smoothing and averaging. (c)–(e) Topological defects and their tracks at different moments of time. Positive and negative defects and their tracks are highlighted in red and blue, respectively. Scale bar is 100 μm . (f) Temporal evolution of the topological polarization calculated as $P = \int_0^L xC(x)dx$, where $C(x)$ is the charge density and $L = 500 \mu\text{m}$ for the presented data. (g) x component (along the activity gradient) of positive (top panel) and negative (bottom panel) defect velocities averaged over y for different x . Orientation of axis is shown in panel (e). The velocity is calculated from the displacement for the previous 0.5 s.

activity in each experiment by extracting the rotational rate of the director field [Fig. 1(b)] averaged over the first few seconds after the AELC is energized, before defects propagate into passive zones. A typical activity distribution is shown in Fig. 2(a) and Movie S3 in the Supplemental Material [30] by saturation of the background (green) color.

The initial distribution of active-passive interfaces in the experimental cell is not correlated with the local director field alignment, and the activity/inactivity interface orientation is often not aligned with the local director field before activity is applied. However, once energized, the emergence of the active-passive interfaces influences the director field. Over time, the director field aligns perpendicular to the activity gradient (i.e., becomes parallel to the interface), as shown in Movie S4 and Fig. S2 in the Supplemental Material [30]. This alignment effect was previously predicted in simulations under conditions of a shallow activity gradient (“weak field”) [20]. In the case of a sharp activity gradient, the theoretical predictions suggest parallel alignment of the director to the activity gradient, a phenomenon that we do not observe experimentally likely due to the relatively broad active-passive interface resulting in a low gradient of activity across the interface.

III. TOPOLOGICAL DEFECT DISTRIBUTION IN A SYSTEM WITH SPATIALLY VARYING ACTIVITY

Upon the application of the acoustic signal, the long-range orientational order within the active region is disrupted.

The interplay between elastic and active forces gives rise to undulation instability [6,31–35], which further evolves with activity into a chaotic motion and proliferation of topological defects (see Fig. 2). In the presence of an activity gradient, the density of topological defects is nonuniform, with higher densities in active zones where defect generation rates are higher [2], as illustrated in Fig. 2(a).

In a typical experiment, the average rotation rate of the director field Ω in the active zone ranges between 0.3 and 0.5 rad/s, and topological defects emerge within 2–5 s after the onset of activity. The behavior of the system is highly uniform along the interface [the y direction in Fig. 3(c)], and the spatial distribution of the defects shows only weak variations with y due to the experimental noise. For this reason, we characterize the defect dynamics solely as a function of x , averaging the defect spatial density over the y direction to improve statistical accuracy. The spatial distribution of defects closely follows the activity profile $\Omega(x)$ for approximately 20 s, as shown in Fig. 3(a). An example of the temporal evolution of the spatial defect profiles for positive defects $D_+(x)$, negative defects $D_-(x)$, and the total defect population $D(x)$ is shown in Fig. S3 of the Supplemental Material [30]. The spatial topological charge density, $C(x) = (D_+(x) - D_-(x))/2$, remains nearly zero, and the active-passive interface does not carry a net topological charge. The net topological charge polarization, defined as $P = \int_0^L xC(x)dx$, fluctuates around zero [see Fig. 3(f)] ($L = 500 \mu\text{m}$ in our experiments).

In active nematic materials with extensile stress generation, positive defects can be considered as active particles

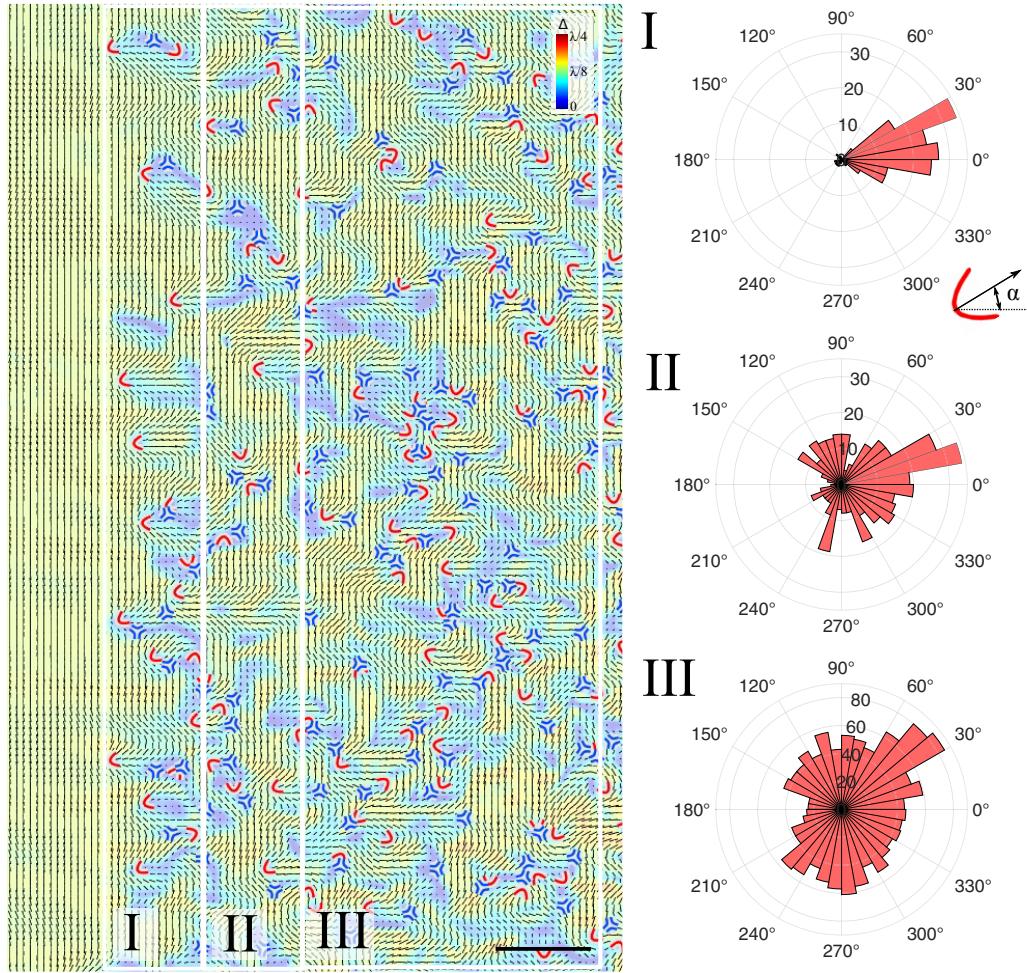


FIG. 4. Orientational order of topological defects. Histograms of the orientation of positive topological defects across different layers near the active-passive interface. In the layer closest to the passive zone, I, the $+1/2$ defects exhibit the strongest alignment, predominantly oriented along the direction of the activity gradient. In the intermediate layer, II, while some defects remain aligned, others are randomly oriented. Deeper within the active region, in layer III, the orientation distribution of defects becomes nearly isotropic. The scale bar is $50\ \mu\text{m}$.

[2,7,8], which are known to accumulate in regions where their motility slows down [8,36,37]. This leads to the clustering and trapping of active particles in passive zones [38–40]. As a result, positive topological defects generated in the active region traverse the active-passive interface and accumulate in the passive zone adjacent to it, as shown in Figs. 3(c) and 3(d) (see also Movies S5 and S6 in the Supplemental Material [30]). The observed behavior is consistent with predictions from recent theoretical studies [8,20].

The x component of the velocity of topological defects averaged over y direction is shown in Fig. 3(g) as a function of time x for different observation times. When activity is on, the topological polarization is driven by the motion of $+1/2$ defects toward the passive zone, while $-1/2$ defects largely remain confined to the active region, with only occasional advection into the passive zone. Consequently, the active-passive interface becomes polarized [see Fig. 3(b) for the topological charge distribution]. Accordingly, the topological charge polarization, P , shifts to negative values as shown in Fig. 3(f). The activity gradient functions like an electric field, inducing a topological charge dipole at the interface by separating defects based on their topological charge [20].

Upon cessation of activity, the topologically charged layer begins to “discharge,” which is manifested by the motion of positive defects in the opposite direction [see Fig. 3(g)]. The defects annihilate in pairs ($+1/2$ and $-1/2$) [see Fig. 3(e)]. The decay follows an approximately exponential form (see the Supplemental Material [30] for the discussion of the exponential behavior). However, the most puzzling behavior occurs within a few seconds immediately following the cessation of activity [between 70 and 80 s in Fig. 3(f)], when the polarization P abruptly drops to even lower values, thereby increasing the overall topological polarization of the layer. This behavior is robust and attributed to the rapid motion of negative defects within the polarized layer toward the positive x direction [see Fig. 3(g) for 70 s], while the positive defects largely remain stationary. The underlying physics behind this behavior remains not fully understood.

IV. TOPOLOGICAL DEFECT ALIGNMENT

Activity gradient in an active nematic medium is predicted to exert a strong aligning effect on positive half-charge topological defects through the mechanism analogous to an

electric field aligning electric dipole moments [8,41,42]. A “weak” activity gradient aligns $+1/2$ defects in a “conventional” way, with the orientation of defects antiparallel to the gradient of activity—analogous to a normal dielectric polarization. This alignment is particularly strong in a layer close to a passive zone (see Fig. 4, zone I), where all defects are aligned. Within the active zone (zone III in Fig. 4), the distribution of defect orientation becomes homogeneous.

In summary, acoustically energized active liquid crystals represent a robust experimental platform for investigating the dynamics of topological defects in active nematic materials. We demonstrate the formation of well-defined activity gradients in the active liquid crystal resulting in the development of active-passive interfaces. The positive topological defects align with the activity gradients and move toward the passive region, leading to the topological charge separation and the formation of a polarized interfacial layer. The strength of the polarization is tuned by activity. Upon cessation of activity, the charge separation at the interface dissipates by topological

defect recombination. The results provide direct experimental validation of theoretical predictions regarding topological defect alignment and charge separation in active nematics with spatial profile of activity. The findings highlight the potential of activity patterning for control and manipulation of topological defect dynamics in active liquid crystals.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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