Direct Observation of the Brownian Motion of a Liquid-Crystal Topological Defect

Chris D. Muzny and Noel A. Clark

Condensed Matter Laboratory, Department of Physics, University of Colorado, Boulder, Colorado 80309

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Optical study of point disclinations in the two-dimensional director field of freely suspended smectic-C liquid-crystal films shows that these defects exhibit Brownian spontaneous positional fluctuations.

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The study of topological defects has played a central role in liquid-crystal (LC) physics since early in the century, providing important clues as to the symmetry and order of various LC phases [1,2]. Among the most useful defects have been the nematic-phase disclination line and point singularities in the orientation of $\mathbf{n}(\mathbf{r})$, the director vector field giving the local mean molecular long axis. These defects are observable via their characteristic signatures in the optical microscope and their static structure [2,3] and deterministic dynamics [2–4] have been widely studied. In this Letter we extend the observation of LC topological defects to include the effects of spontaneous thermal fluctuations, studying the spontaneous motion of a single 2π point disclination in the two-dimensional (2D) director field of an ultrathin freely suspended smectic-C liquid-crystal film. We show that, as suggested by Van Winkle and Clark [5], a 2D 2π disclination behaves like a particle executing Brownian motion. We find the diffusion constant for the defect and compare our result to those predicted by Pleiner [6], using a continuum elastohydrodynamic model, and to those of Loft and DeGrand [7] found using an XY-model molecular-dynamics computer simulation.

Smectic C is a layered liquid-crystal phase having the mean molecular long axis, given by the director $\mathbf{n}$, tilted relative to the layer normal through an equilibrium angle θ [3]. Smectic C's readily form few-layer-thick freely suspended films [8] in which the projection of $\mathbf{n}(x,y)$ onto the film plane defines a two-dimensional vector field $\mathbf{c}(x,y)$, which can be completely described by its scalar azimuthal orientation $\phi(x,y)$ [8], and can be visualized for experimental study using depolarized reflected light microscopy (DRLM) [9]. In the current experiment the circular nature of the film holder together with a strong pinning boundary condition fixing the orientation of $\mathbf{c}$ relative to the edge of the holder combined to produce a single $+2\pi$ $(s=+1)$ point disclination having the $\mathbf{c}(x,y)$ structure in Fig. 1(a), appearing in DRLM as the characteristic four-arm brush shown in Fig. 1(b) [9]. The feathery texture of the brushes is a result of finite-amplitude orientation (spin-wave) fluctuations in $\mathbf{c}(x,y)$, which have been studied by light scattering [8,10] and cross-correlation techniques [11].

Films were drawn in the smectic-A phase across a 3.7-mm-diam hole in a microscope cover slip, the hole having been polished to reduce distortions in the director field due to irregularities at the edge. As will be discussed below, it was desirable to use films which were as thin as possible in order to maximize the defect diffusion constant. The experiments, using the material 10.O7* [12], were therefore done with three-layer films ($t \cong 75$ Å, determined by optical reflectivity), the minimum readily obtainable thickness for this material. Films were contained in a temperature-controlled microscope oven with 1-mK stability [11] and DRLM with incident 4880-Å laser light used to visualize $\mathbf{c}(x,y)$. The DRLM image was focused onto a Vidicon video camera and recorded on a VHS videotape recorder with a 30-Hz frame rate. The taped images were then digitized on a Sun-based image processing system.

The defects show up in DRLM as shown in Fig. 1(b) and from analysis of the image versus analyzer orientation it was determined that they were bend defects as in Fig. 1(a). The brush intersection produced a well-defined fourfold-symmetric pattern in its vicinity, the center of which was taken to be the defect core location in a given frame. The trajectory of defects was tracked at two different temperatures in the smectic-C phase, 74.6 and 55.1 °C. The results for the two cases were significantly different and will be presented separately. Figure 2(a) shows a sample trajectory of the defect over a 2-h period at 55.1 °C and Fig. 2(b) is a plot of the corresponding mean-square displacement $\langle \Delta r^2 \rangle$ versus time t, where Δr

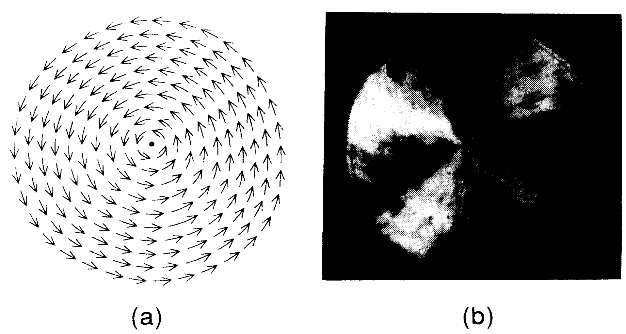


FIG. 1. (a) The topological defect studied in these experiments: a 2π $(s=+1)$ point bend disclination in $\mathbf{c}(\mathbf{r})$, the 2D nematic director field of a freely suspended smectic-C film. (b) Appearance of the vortex of (a) in depolarized reflected light microscopy (DRLM) [9]. The diameter of the illuminated area is 1.8 mm. The film diameter is 3.7 mm.

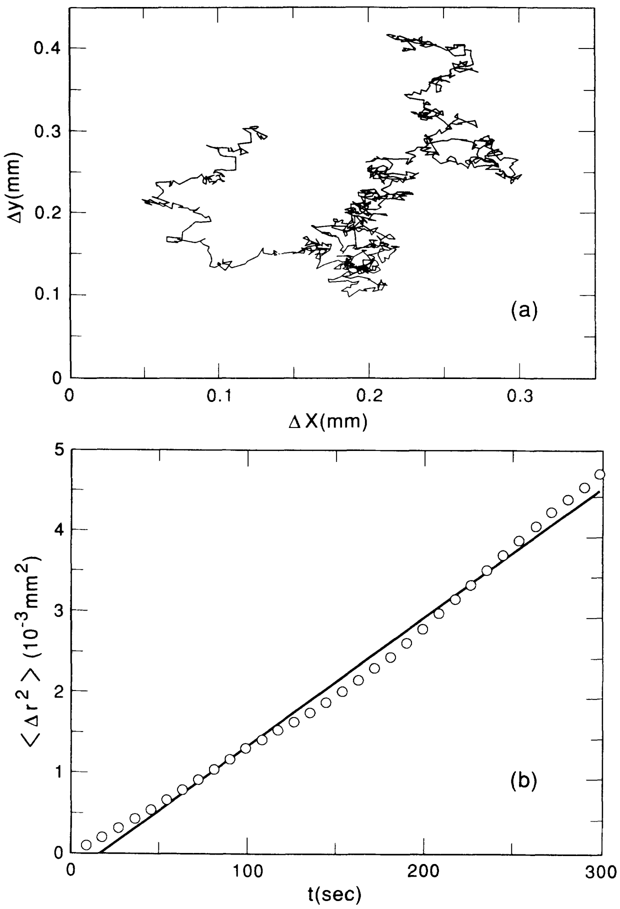


FIG. 2. (a) The trajectory of a defect over a 2-h time period at 55.1 °C. (b) Mean-square displacement vs time for the data in (a), giving a diffusion constant $D=0.8\times10^{-7}$ cm^2/sec.

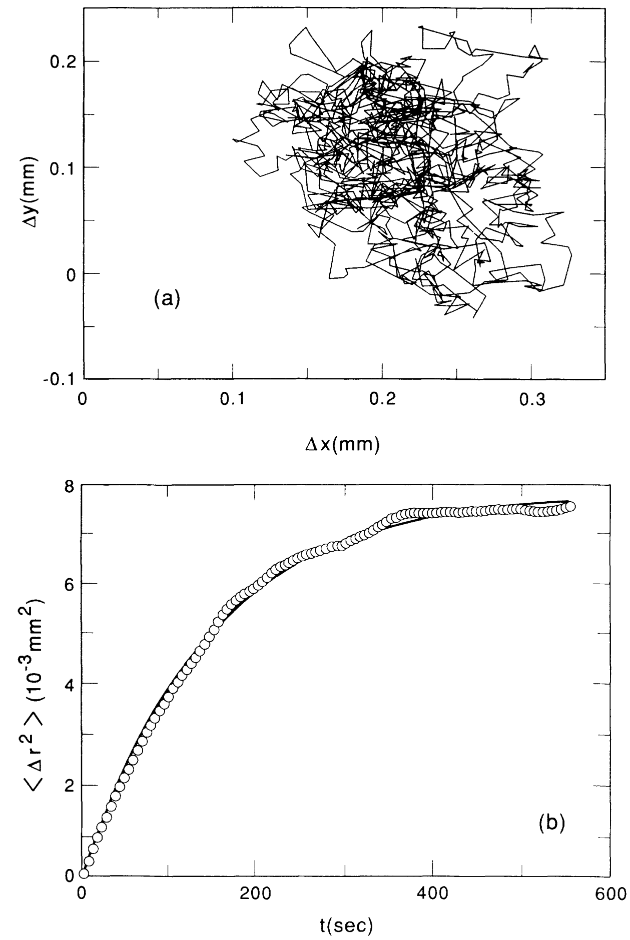


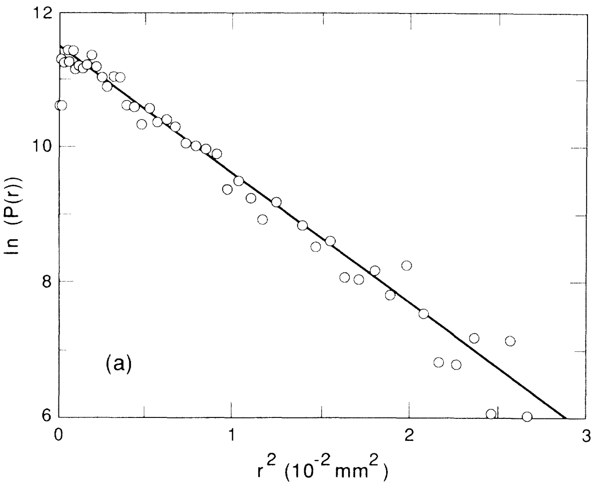
FIG. 3. (a) The trajectory of a defect over a 2-h time period at 74.6 °C. (b) Mean-square displacement vs time for the data in (a). The saturation of $\langle\Delta r^2\rangle$ at long times shows the effect of the binding potential on the motion.

is the defect location relative to its starting position at $t=0$. As can be seen, $\langle\Delta r^2\rangle$ is linear in t, indicating Brownian-like diffusion of the defect. This defect is diffusing slowly and, over the period of the experiment, the effects of the potential well are not noticeable. The diffusion constant obtained from the slope of $\langle\Delta r^2\rangle$ vs t is $D=0.8\times10^{-7}$ cm^2/sec. Figure 3(a) shows a 2-h trajectory of the defect at 74.6 °C and Fig. 3(b) the corresponding mean-square displacement versus time. Qualitatively, by comparing to Fig. 2(a), it can be seen that the defect is diffusing much more rapidly than it did at the lower temperature and that this trajectory is bound to an approximately circular region near the center of the film holder, with $\langle\Delta r^2\rangle$ vs t initially increasing linearly but then saturating as a result of the confining potential. The saturation indicates that the defect has achieved equilibrium in the confining potential. We can then assume a Boltzmann radial probability distribution of defect positions, $P(r)$, for the potential $U(r)=Cr^2/2$, justified below, for which the slope of $\ln P(r)$ vs r^2 in Fig. 4(a) is just $C/2k_BT$, giving $C=1.8\times10^{-9}$ erg/cm^2. From the initial slope of $\langle\Delta r^2\rangle$ vs t in Fig. 3(b) we determine the diffusion constant $D=1.8\times10^{-7}$ cm^2/sec, as shown in Fig. 4(b). This value is twice that at the lower temperature.

We thus have presented our basic experimental result, namely, that point disclinations in freely suspended films exhibit Brownian positional fluctuations. We now discuss this result in the context of available models. The static behavior of such a 2D point defect is described by Frank elasticity. For a film of thickness t the free energy of the field $\phi(x,y)$ in the one-elastic-constant approximation is just that of the 2D XY model [3,8,13],

$$F=\frac{1}{2}\int\{K_n t\sin^2\theta\}(\nabla\phi)^2dx\,dy$$
$$\equiv\frac{1}{2}\int\{K\}(\nabla\phi)^2dx\,dy\,, \qquad (1)$$

where K is the effective 2D Frank elastic constant and K_n is the 3D nematic Frank elastic constant in the one-constant approximation. This free energy, when combined with the boundary condition that **c** is locally paral-



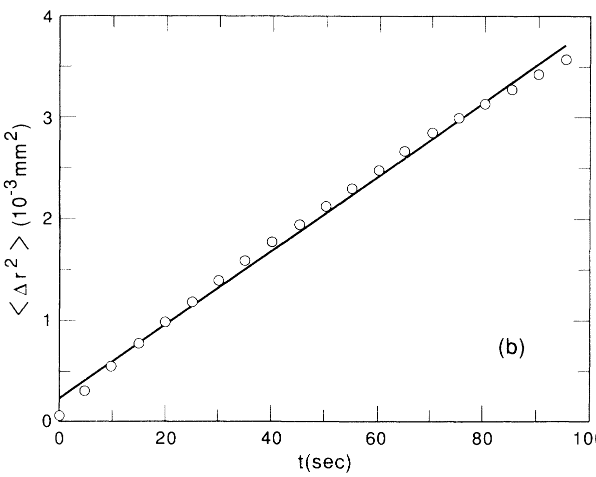


FIG. 4. (a) A ln plot of $P(r)$, the probability of displacement r of the defect from the film center vs r^2 for the data of Fig. 3(a). The slope gives the elastic constant C for the binding potential. (b) Short-time mean-square displacement vs time for the data of Fig. 3(a), giving $D=1.8\times10^{-7}$ cm^2/sec.

lel to the edge of a circular film holder of radius R, gives the thermal-fluctuation-free solution for a single $s=+1$ defect on a circular film [5,6]:

$$\phi(\mathbf{r})=\tan^{-1}[y/(x-x_0)]+\tan^{-1}[y/(x-R^2/x_0)] .$$

Here $\mathbf{r}=0$ is at the hole center and the defect is at $(x_0,0)$. From this form the energy $U(r)$ of the defect versus displacement r from the film center is found to be $U(r)=Cr^2/2$, that is, the defect is harmonically bound to the hole center with a spring constant C, where [5,6]

$$C=2\pi K/R^2=2\pi\{K_n t\sin^2\theta\}/R^2 . \qquad (2)$$

The mean-square defect displacement from center will be $\langle r^2\rangle=k_BTR^2/2\pi K\sim R^2a/t\sim R^2/n$, where a is a molecular length and n is the number of layers in the film. From our measured value of C and Eq. (2) we find the bend elastic constant for the film to be $K=9.7\times10^{-12}$ erg, which is comparable to K obtained via orientation fluctuation spectroscopy in similar systems [8,10].

Pleiner [6] has considered the solution of the nematodynamic equations for the static structure, deterministic motion, and diffusive motion of the $s=+1$ defect. He considers a defect core of radius r_c which is smectic-A-like ($\theta=0$), developing an equation of motion for the defect core coordinate $\mathbf{r}(t)$, which is just the Langevin equation for a single particle: $m_{\rm eff}d^2\mathbf{r}/dt^2+\mu\, d\mathbf{r}/dt+C\mathbf{r}=m_{\rm eff}\xi(t)$. Here $m_{\rm eff}$ is the defect effective mass, $\mu=v/F$ is the defect mobility (velocity per unit applied force), and $\xi(t)$ is a fluctuating force with $\langle\xi(t)\xi(t')\rangle=\Gamma\delta(t-t')$. $m_{\rm eff}$ comes from the kinetic energy associated with the change of θ as the core moves, and μ comes from the reorientation of $\mathbf{c}$: As the defect moves by, the molecules continually reorient in ϕ. Pleiner in calculating μ includes the coupling to flow of the film but shows that backflow effects are negligible. The mobility is then obtained by integrating the local dissipation, $\gamma(d\phi/dt)^2=\gamma_n t\sin^2\theta(\mathbf{v}\cdot\mathbf{r}/r^2)^2$ over the film area and equating it to v^2/μ. The result for μ is [5,6]

$$\mu^{-1}=2\pi\gamma\ln(R/r_c) , \qquad (3)$$

where $\gamma=\gamma_n t\sin^2\theta$ is the effective 2D nematic orientational viscosity and γ_n is the 3D nematic orientational viscosity. The fluctuating force $\xi(t)$ is obtained from equipartition, i.e., the coordinate $\mathbf{r}(t)$ is assigned a *single* degree of freedom. The Langevin equation contains two characteristic times, the defect velocity correlation time $\tau_v=\mu m_{\rm eff}=(\gamma/8K)r_c^2\ln(R/r_c)$ (the orientational diffusion time for fluctuation of dimension r_c), and $\tau_r=1/\mu C$, the deterministic relaxation time for return to equilibrium if the defect is displaced from the film center. The core radius can be estimated from typical smectic-A to -C transition Landau parameters [14] to be 100 Å $<r_c<1000$ Å, for which τ_v is in the microsecond regime. Thus, with the $\frac{1}{30}$-sec time resolution of the experiment, diffusive motion is predicted in this model and, as shown above, observed. The calculated diffusion constant is $D=k_BT\mu$ which, with the above estimate for r_c and viscosity measured in similar systems [10], gives $D\sim0.5\times10^{-7}$ cm^2/sec, in reasonable agreement with the experiment. A quantitative test of the model awaits determination of the elastic constants and viscosities for 10.O7*, and experiments involving both diffusive and deterministic motion, enabling independent determination of μ and D.

We have also compared our diffusion constant to that measured in an XY-model molecular-dynamics computer experiment [7]. This experiment gives $D_{XY}=1.81$ in units of $D_s=K/\gamma$, the spin-wave diffusion constant. Using typical values for D_s [10,11] we find $D_{XY}=1\times10^{-6}$ cm^2/sec, considerably larger than either our measured values or the continuum model.

We now consider briefly the possibility of observing thermal fluctuations of three-dimensional nematic disclinations. Because of their excess internal energy 3D disclination lines are effectively under a tension which has the value of a typical nematic Frank constant, $K \sim k_B T / a$. A simple calculation shows that the local mean-square fluctuation in position of a line of length R in 3D will be $\langle u^2 \rangle = Ra$ which is not resolvable unless R is ~ 1 cm or greater. By using the free film we effectively take a slice of the nematic of thickness t, giving $\langle u^2 \rangle = Ra(R/t)$ enhancing $\langle u^2 \rangle$ by the large ratio R/t so that fluctuations are readily observable. Point disclinations in nematics are often found at surfaces but are trapped by surface defects. It may be possible to observe fluctuation effects for defects on free (air-LC) interfaces.

In conclusion, we have observed directly the spontaneous thermal motion of a topological defect, finding that a 2π XY vortex in the nematic director field of a freely suspended smectic-C liquid-crystal film behaves like a Brownian particle in a potential well. The thermal motion of this defect is large and easily observable by optical microscopy. By analyzing defect position versus time we were able to determine diffusion constants at two different temperatures and found that the predictions of Pleiner's continuum elastohydrodynamic model agree well with our observations.

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[12] 10.O7* is the compound 4-[(S)-(4-methylhexyl)oxy]-phenyl 4-(decyloxy)benzoate, first reported by P. Keller, Ferroelectrics **58**, 3 (1984). Its phase sequence in bulk is

$$I \xrightarrow{80^\circ} N^* \xrightarrow{76^\circ} \text{Sm-}A \xrightarrow{73^\circ} \text{Sm-}C^* \xrightarrow{43^\circ} \text{Sm-}X \xrightarrow{35^\circ} X\,.$$

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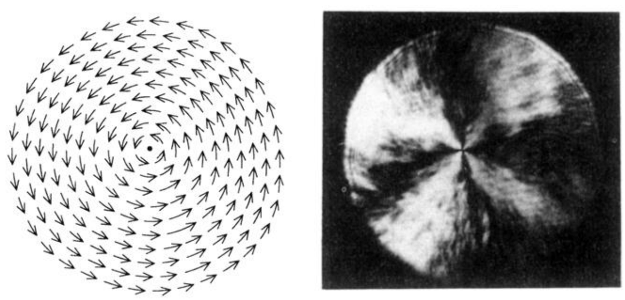

(a) (b)

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