Dynamics of Smectic-C Point Disclinations in Freely-Suspended Liquid Crystal Films

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Dynamics of Smectic-C Point Disclinations in Freely-Suspended Liquid Crystal Films

by

Kathryn Wachs

A thesis submitted to the faculty of the University of Colorado in partial fulfillment of the requirements for the award of departmental honors in the Department of Physics

2014
This thesis entitled:
Dynamics of Smectic-C Point Disclinations in Freely-Suspended Liquid Crystal Films
written by Kathryn Wachs
has been approved for the Department of Physics

Joseph Maclennan

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The final copy of this thesis has been examined by the signatories, and we find that both the content and the form meet acceptable presentation standards of scholarly work in the above mentioned discipline.
Wachs, Kathryn (B.A., Physics)

Dynamics of Smectic-C Point Disclinations in Freely-Suspended Liquid Crystal Films

Thesis directed by Prof. Joseph Maclennan

The motion of topological defects in the director field is observed in freely-suspended smectic-C liquid crystal films using reflected polarized light microscopy. We study the annihilation of pairs of defects with opposite topological charge and observe the decreasing separation of +1 and -1 defects as a function of time, which we compare with available theories. We measure a relative increase in stochastic influences as the defects near coalescence. We also observe the diffusive motion of single point disclinations. We study the effect of surrounding air pressure on the mobility of a single +1 defect. We find that lowering the pressure leads to an increase in the diffusion coefficient. At sufficiently low pressure, the defects disappear, which suggests a transition to the smectic-A phase.
Acknowledgements

I owe several people thanks in the creation of this work. The first of which is Joseph Maclennan, without whom I would not have this job. His calming insistence to always go back to the basics and use real nouns, along with his inclusion of me, were of considerable help in the creation of this piece. Cheol Park, whose patient aid and good humor taught me how to actually conduct experiments. Noel Clark, my conversations with whom always pointed out the big picture of what I was doing and why. Arthur Klittnick, who can find any piece of equipment and never said no to a confused undergrad. I would also like to thank my labmates, Zhiyuan Qi, whose enthusiasm and vacuum setup practically started one of my projects, and Kyle Meienberg, who always kept me honest and made lab life more entertaining. Finally, I would like to thank everyone in the group who accepted me as one of their own.
Contents

Chapter

1 Liquid Crystals, Smectics, and Their Observation 1
   1.1 Liquid Crystals and Smectics ............................................. 1
   1.2 Defects and Islands ...................................................... 3
   1.3 Observation .............................................................. 6
   1.4 Software ................................................................. 8

2 Dynamics of Oppositely-Charged Coalescing Topological Defect Pairs 10
   2.1 Experiment ................................................................. 10
   2.2 Theory ................................................................. 11
   2.3 Results ................................................................. 13
   2.4 Conclusion ............................................................. 18

3 Effects of Surrounding Air Pressure on Single Defect Diffusion 19
   3.1 Experiment ................................................................. 19
   3.2 Theory and Previous Experiments .................................... 20
   3.3 Results ................................................................. 20
      3.3.1 Mobility and Decreasing Pressure .............................. 20
      3.3.2 Defect Disappearance ........................................... 24
   3.4 Conclusion ............................................................. 24
# Figures

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1</td>
<td>Schematic representation of solid, LC, and liquid phases.</td>
<td>2</td>
</tr>
<tr>
<td>1.2</td>
<td>Schematic representation of SmA and SmC phases.</td>
<td>3</td>
</tr>
<tr>
<td>1.3</td>
<td>Defects in a SmC film.</td>
<td>4</td>
</tr>
<tr>
<td>1.4</td>
<td>Island boundary conditions.</td>
<td>6</td>
</tr>
<tr>
<td>1.5</td>
<td>Freely-suspended film diagram.</td>
<td>7</td>
</tr>
<tr>
<td>1.6</td>
<td>Reflected polarized light microscopy setup.</td>
<td>8</td>
</tr>
<tr>
<td>1.7</td>
<td>Tracking program procedure.</td>
<td>9</td>
</tr>
<tr>
<td>2.1</td>
<td>Chemical structure of 3M900084.</td>
<td>11</td>
</tr>
<tr>
<td>2.2</td>
<td>Sample experimental image of defect annihilation.</td>
<td>12</td>
</tr>
<tr>
<td>2.3</td>
<td>Defect positions.</td>
<td>13</td>
</tr>
<tr>
<td>2.4</td>
<td>Defect separations as a function of time.</td>
<td>13</td>
</tr>
<tr>
<td>2.5</td>
<td>Defect separations and their fits.</td>
<td>14</td>
</tr>
<tr>
<td>2.6</td>
<td>Defect comparing separations.</td>
<td>15</td>
</tr>
<tr>
<td>2.7</td>
<td>Average defect separation velocity as a function of separation.</td>
<td>15</td>
</tr>
<tr>
<td>2.8</td>
<td>Mutual displacement fluctuations as a function of separation.</td>
<td>16</td>
</tr>
<tr>
<td>2.9</td>
<td>Average velocity times separation compared to theory.</td>
<td>17</td>
</tr>
<tr>
<td>2.10</td>
<td>Experimental defect orientations.</td>
<td>17</td>
</tr>
<tr>
<td>2.11</td>
<td>Possible defect orientations.</td>
<td>18</td>
</tr>
</tbody>
</table>
3.1 The vacuum setup. ................................................................. 20
3.2 Previous inclusion experimental results. ................................. 21
3.3 Diffusion coefficient versus air pressure for a defect. ................. 22
3.4 Probability of defect position as a function of separation. ......... 23
3.5 Elastic constant of the film as a function of pressure. ............... 23
Chapter 1

Liquid Crystals, Smectics, and Their Observation

Liquid crystal (LC) systems are ordered fluids that exhibit non-trivial physics. They have electro-optic properties that allow them to be easily observed [31, 8], and so are widely-used in the display industry in the form of liquid crystal displays [18]. Thin smectic LC films are ideal examples of two-dimensional (2D) fluids, because although they are typically several mm in diameter, they comprise just a few layers, each a molecule (or less) thick [31, 8]. Optical microscopy can be used to study broken symmetries of smectic-C films [8, 28], exemplified by point disclinations called defects. These defects are characteristic of broken symmetry systems also found in other areas of physics such as cosmology, particle physics, and condensed matter physics [28]. Smectic-C defects have been studied and modeled in the past [23, 16, 12, 9], but our research suggests existing models may be incomplete.

This thesis is organized as follows. In Chapter 1, we will discuss relevant background information on smectic LCs and their observation and analysis. In Chapter 2, we will investigate the annihilation dynamics of two isolated, oppositely topologically charged defects. Chapter 3 will examine the Brownian diffusion of single +1 defects with decreasing ambient air pressure.

1.1 Liquid Crystals and Smectics

Liquid crystal phases lie between the solid and liquid phases. LC molecules do not have random orientation, as in liquids, nor rigid positional and orientational order, as in solids. Instead, they have a predilection for a certain orientation like solids with the fluidity of a liquid (see Fig.
All types of LCs have orientational order, and a few have positional order. Thermotropic LCs are organic compounds that change phase according to temperature. These are contrasted with lyotropic LCs which depend on temperature and concentration in a solvent. The molecules in most LCs are generally either rod-like (calamitic LCs) or disc-like (discotic LCs). In either case, the molecules have an average orientation described by the \( \mathbf{n} \)-director, as in Fig. 1.1(b) [19].

Calamitic LCs have several possible subphases: nematic, chiral nematic, and smectic. In the nematic phase, the molecules are roughly parallel to one another, having orientational but no positional order. In the chiral nematic phase, the \( \mathbf{n} \)-director continually rotates throughout the sample, with the molecules twisting along one direction with a temperature-dependent pitch [19].

In the smectic LC phase, the molecules have additional positional order and arrange into discrete layers. Since smectic films have integer layer thickness and the intensity of light reflected from the film is proportional to the square of its thickness [14], the number of layers can be easily determined experimentally. Two common smectic phases are smectics-A and -C. In the smectic-A (SmA) phase, the molecules are oriented on average perpendicular to the layer plane, while in the smectic-C phase (SmC), the molecules are tilted from the layer normal. The azimuthal orientation of the molecules in the SmC phase may be described by the projection of the \( \mathbf{n} \)-director into the layer plane, called the \( \mathbf{c} \)-director. The \( \mathbf{c} \)-director is commonly depicted as a nail, the head of which corresponds to the end of the molecule closest to the viewer, as in Fig. 1.2(b) [19]. Because the films we study are very thin (~6 nm), their aspect ratio is around 1:10\(^6\), and they are model 2D systems [31].
A chiral molecule is one where the mirror image is not superimposable on the original. These molecules can be either right- or left-handed, according to the molecular conformation. Chiral smectic-C films (SmC*) have spontaneous polarization perpendicular to the c-director and in the layer plane. Applying an electric field to a chiral material will cause the molecular dipoles to align with the field. Racemic materials have half right- and half left-handed molecules. Achiral materials have no chirality and the mirror images of the molecules are superimposable over the original [27].

The free energy density of a SmC film may be described in terms of c-director distortions. The Frank elastic energy can be approximated as

$$F = \frac{1}{2} \int \left[ K_s^{2D} (\nabla \cdot \mathbf{c})^2 + K_b^{2D} (\nabla \times \mathbf{c})^2 \right] d^2 r,$$

where $K_s^{2D}$ is the 2D splay Frank elastic constant and $K_b^{2D}$ is the 2D bend Frank elastic constant. In our equations, these two elastic constants are taken to be equal [18]. One can see from Eq. 1.1 that the c-director field in a SmC film is preferentially uniform, with the same c-director throughout. In the lowest energy state of the film, the thickness is also uniform [22]. However, when films are perturbed, they exhibit non-trivial physics that is characteristic of broken symmetry systems. The most common such phenomena are defects and islands [23, 20, 8].

1.2 Defects and Islands

A point disclination is a topological defect within the film where the n-director points directly upward. As in the SmA case, there is no c-director at that point. Other topological defects include...
walls and hairpin defects, more common in other types of LCs. Point defects act as topological charges, with the charge designated by how much the director changes in a counter-clockwise circle around the defect, and defects with opposite charges attracting. For instance, for a +1 defect, the director will change by $2\pi$ in a counter-clockwise circle around it. However, for a $-1$ defect the director changes by $-2\pi$. That is, $\Delta\Phi/\Delta\phi = \pm 1$ for strength 1 defects, where $\Delta\Phi$ is the angle subtended as you move around the defect and $\Delta\phi$ is the azimuthal rotation of the director around the defect. Examples of both defects are shown in Fig. 1.3. The defect itself is the point at the center of each image where the $c$-director has a singularity [16, 8]. Defects occur spontaneously after disturbing the film or are created in a circular film or an island due to boundary conditions constraining the $c$-director at the edges.

SmC defects undergo Brownian motion, i.e., motion due to random collisions of the molecules within the film. In Brownian motion, the mean-squared displacement, $\langle \Delta r^2 \rangle$, increases linearly as a function of time, with a slope proportional to the diffusion coefficient, $D$, and the mobility, $\mu$ [12]. However, the exact manner in which defects move, whether as a core radius of molecules moving as one with the other molecules reorienting around them, or as only a reorientation of the molecules, is still an open area of investigation, one we will further discuss in Chapter 3.

![Image of defects](image)

Figure 1.3: Defects in a SmC film. (a) Image of a +1 defect, viewed in reflected light under polarizer and analyzer crossed vertically and horizontally. Schematic representations of (b) a +1 and (c) a $-1$ defect, adapted from [8]. $\Phi$ is the angle around the defect, while $\phi$ is the angle of the $c$-director. The $c$-director is represented by a nail, where the head is the end of the molecule closest to the viewer. The defect is the point in the center of these images where the $c$-director does not exist.
Because defects are essentially massless, they theoretically should have infinite mobility, but they are “locked in” due to the elastic energy constraints of the film. For example, since the glass film holder exhibits a boundary condition on the c-director, the defect can only move a certain distance from the film center before the energy cost, which depends on the orientational gradients, becomes too high. Earlier work has shown that in a SmC film of finite size, while defects undergo Brownian motion at shorter time scales, for longer ones, the mean-squared displacement of defects saturates. Defects are, in a sense, in a potential well, whose shape depends on the elasticity of the film [12]. Defect speed is limited by the viscosity of the film, or the dissipation associated with director reorientation, and by the viscosity of the surrounding medium, which exerts a drag force [23].

Islands are topological inclusions consisting of more layers than the surrounding film. The island edges, which are dislocations, impose a strict boundary condition on the relative orientations of the molecules near them. Because of these boundary conditions, a +1 defect forms within the center of each island. To maintain a net topological charge of zero, an accompanying −1 defect forms in the film outside the island, as indicated in Fig. 1.4. The island and −1 defect move like a dipole, with the motion of the −1 defect constrained according to the motion of the island [26, 20]. Both defects as well as the island undergo Brownian motion [14]. For both the island and the defect, the time it takes for the film to “forget” which way it was going before is small enough that we can assume it goes in a new, random direction at each observed time step, as in normal Brownian motion [27].

The motion of a single inclusion in a 2D fluid has been accurately modeled by SD-HPW theory. While the influence of an inclusion on the surrounding fluid falls off as $1/R$ in 3D, that of a inclusion on a 2D fluid falls off logarithmically. These differences arise from the solution to Laplace’s Equation in two and three dimensions. An inclusion in an LC film acts as a particle in a quasi-2D system: a 2D film surrounded by a 3D viscous medium. The distance to the 2D-to-3D boundary, i.e., the circle around the inclusion within which the velocity field falls off logarithmically and beyond which it falls off as $1/R$, is defined as the Saffman length, $l_s$. This length depends
on the viscosity of the film and that of the surrounding medium. To a good approximation, the fluid inside the Saffman length may be thought of as moving with the inclusion, since logarithmic decay is so much slower than $1/R$. The model was developed first by Saffman and Delbrück [15], expanded upon by Saffman [25], and finally generalized by Hughes, Pailthorpe, and White (HPW) [5]. The SD-HPW theory was experimentally verified for islands diffusing in SmA films [14].

1.3 Observation

Freely-suspended films of smectic LCs are near-2D systems [31]. Smectic material is spread on a film holder, which consists of a glass cover slide with a hole cut out of it. This is laid over a piece of metal with another hole in it for easy maneuverability and to prevent the shattering of the glass cover slide. The film is spread across the opening using a glass cover slide known as a “spreader,” as shown in Fig. 1.5. Drawing a two-layer film is a delicate process, necessitating drawing a thick film with a bilayer “hole”. These appear darker than the surrounding material and therefore look like holes in the film. Such holes expand at varying speeds to take up the entirety of the film and impose boundary conditions on the director that induce a $+1$ defect at the hole center. As holes grow, excess material from the meniscus often forms islands. Consequently, it is difficult to create a uniform bilayer film over the entirety of the film.
Topological defects are discontinuities in c-director uniformity, and as such are visible only in polarized light. LCs are birefringent materials, meaning that the index of refraction is different for light of different polarizations. If the incident light is polarized parallel or perpendicular to the optic axis, the light will not change polarization. However, if the incident beam has any other polarization, the material changes the polarization of the light, in this case to be elliptically polarized. Under crossed polarizers (i.e., a polarizer and an analyzer at 90°), as in Fig. 1.3(a), the defect looks like the center of a Maltese cross, with four dark and four light brushes. This is because the analyzer does not transmit light polarized perpendicular to the direction of its polarization [18].

In our experiments, films are observed using reflected polarized light microscopy. This is accomplished by streaming the light through a polarizer and focusing lens, down through the objective to strike the film at oblique incidence. The light is reflected from the film back through the objective, through another focusing lens and an analyzer, shown in Fig. 1.6 [8]. The intensity of reflected light is proportional to the square of the number of layers within the film, so islands appear brighter than the surrounding film. For films with more than 20 layers, the number of layers also affects the spectrum of the reflected light [22].
1.4 Software

The defects and islands are tracked using an internally developed Interactive Data Language (IDL) program called MultiTrack. An example of the tracking procedure is shown in Fig. 1.7.

For defects, the program takes an initial coordinate estimate from the user and creates a search box around that location. It also creates a defect template that contains either two or four brushes, allowing the user to track defects in data sets taken under uncrossed or crossed polarizers, respectively. This template is cross-correlated with the image at all orientations and positions within the search box. The peak of this cross-correlation is fitted with a two-dimensional Gaussian to determine the defect position.

Islands and holes may be tracked using the same IDL routine. Island edges are found utilizing the Canny edge detection method [1]. An initial estimate of the edge location is given by the user, and the program then searches a given distance away from the estimate in either direction. It fits
the edges found in that region to a circle, since all islands, and most holes, are circular. The center and radius of this circle are recorded for each video frame.

![Figure 1.7: Tracking program procedure. An image of a defect viewed with decrossed polarizers (a) is cross-correlated with a defect template (b) to find the defect position (c). The hole or island edges are found using edge detection (d) and the island center is determined by fitting a circle to the boundary (e).](image)

Image contrast is increased by translating the images to black and white and performing some image processing. For example, the images can have a pedestal subtracted from them, as well as thresholding. More importantly for old data, it has various noise reduction options, including smoothing using the IDL built-in smooth procedure or a weighted average.

The diffusion constant of a defect or island is measured using a Python program created by Zoom Nguyen [14]. This program determines the mean-squared displacement ($< \Delta r^2 >$) of the defect over time, subtracting off any drift. The slope of the $< \Delta r^2 >$ vs. time plot is proportional to the diffusion coefficient, $D$. 


Chapter 2

Dynamics of Oppositely-Charged Coalescing Topological Defect Pairs

We first examine the annihilation of pairs of defects of opposite strength in two-layer smectic-C liquid crystal films. To minimize the elastic energy of the film, defects of opposite topological charge are attracted to one another. However, there are hydrodynamic drag forces that oppose this attraction and flow fields make the system more complicated [28, 32]. Experiments were carried out by former graduate student Darren Link several years ago and analyzed by me to determine the dynamics of defect coalescence.

2.1 Experiment

Freely-suspended films were created and observed using polarized reflected light microscopy. To avoid vibrations, thermal fluctuations, and air currents causing excess noise in the data, these films were drawn inside a temperature-controlled oven. The oven was composed of two large aluminum pieces attached to a heating element, thereby creating a contained area with constant temperature while simultaneously providing enough maneuverability for the drawing and quenching of films. The film holder had a 4 mm diameter hole and the spreader was mounted on a push rod within the oven. The films were observed on a rotating x-y stage under a Zeiss Ultraphot photomicroscope [8].

The film was observed under crossed polarizers as shown in Fig. 1.6. A Hitachi RGB camera (model HVC-20) was fitted to the microscope and movies of the experiment were recorded on a Sony Betacam videocassette recorder (model UVW-1400A) [8]. The material used was 3M900084,
an achiral liquid crystal, shown in Fig. 2.1. The two-layer films were studied at \( \sim 76^\circ\text{C} \) and 29.97 frames per second. 3M900084 has the following phase sequence [8]: Crystal \( \rightarrow \) (71°C) \( \rightarrow \) SmC \( \rightarrow \) (79°C) \( \rightarrow \) SmA \( \rightarrow \) (136°C) \( \rightarrow \) Isotropic

![Chemical structure of 3M900084.](image)

Figure 2.1: Chemical structure of 3M900084.

Topological defects are induced via quenching: less than 10 \( \mu \text{L} \) of air is slowly pumped under the film to deform it, inducing the SmA phase. The air is then quickly released, allowing the film to relax back into planar SmC. The molecules that were on average pointing parallel to the layer normal then fall into their SmC orientations more or less at random, inducing defects. The number of defects produced is proportional to the volume of air pumped in and the rate at which the air is released. Islands are induced if the initial film deformation is too large, while too small a deformation fails to induce any defects. The quench, in its entirety, takes place in about 30 ms, or one frame of the video camera. After a short time, typically less than a minute after the quench, only a single pair of oppositely-charged defects remains. We analyze the motion of this last annihilating pair.

2.2 Theory

The simplest model of the dynamics of a pair of annihilating defects assumes a single elastic constant and considers the attractive force, \( F_a \propto \frac{K}{s} \), where \( K \) is the elastic constant and \( s \) is the separation of the defects, and the fluid drag force \( F_f \propto -\eta v \), where \( \eta \) is proportional to the viscosity and the relative velocity \( v \equiv \frac{\partial s}{\partial t} \). Balancing these forces, we obtain

\[
sv \propto -\frac{K}{\eta}
\]  

(2.1)
Figure 2.2: Sample image from experimental data of a pair of annihilating defects in a two-layer film of 3M900084. The width of this image is 1.1 mm.

and

\[ s \propto (t_0 - t)^{1/2}. \]  \hspace{1cm} (2.2)

This model of defect coalescence behavior will be referred to as the square root model.

In 1993, Yurke et al. [32] modified the simple square root model by adding a logarithmic term arising from the drag force that depends on the radius of the defect. The Yurke model gives

\[ s^2 \ln \left( \frac{s}{r_c} \right) \propto t_0 - t \]  \hspace{1cm} (2.3)

or

\[ \frac{1}{s \ln \left( \frac{s}{r_c} \right)} \propto \frac{\partial s}{\partial t}, \] \hspace{1cm} (2.4)

where \( r_c \) is the core radius of the defect.

In 2003, Svenšek and Žumer [28] modeled hydrodynamic and elastic anisotropy effects within SmC films numerically and observed an asymmetry in the defect motion. The +1 defect moves faster than the -1 defect, resulting from the interference of the viscous flow fields and from the elastic anisotropy. This anisotropy is most pronounced in chiral materials, which have spontaneous polarization. Their model suggests that the defects should coalesce faster than the analytic theory of Yurke et al. predicts.
2.3 Results

We extracted images from movies of 32 different annihilation events and used the MultiTrack procedure described in Chapter 1.4 to track the positions of the topological defects, with selected results shown in Fig. 2.3. We then calculated the separation \( s(t) \), the average velocity, and the mutual displacement fluctuations of the defects as they annihilated, as shown in Figs. 2.4, 2.7, and 2.8 respectively.

![Figure 2.3: Two examples of the trajectories of +1 and -1 defect pairs as they diffuse and annihilate.](image)

![Figure 2.4: Two examples of the separation \( s(t) \) between the +1 and -1 defects as they diffuse and annihilate.](image)

The separation versus time data were fitted to both the square root model and the Yurke model with varying degrees of success. In all instances, the models fit better at larger distances.
than at shorter ones. The best fits of selected data sets are shown in Fig. 2.5. For most events, one of the two models may provide a better fit at small separations, but on average, both fits work just as well. With the majority of events, annihilation was observed slower than predicted by the square root model. Annihilation was also experimentally slower than the prediction of Svenšek and Žumer, which stated that flow effects should cause the defects to proceed to annihilation even faster than predicted by the square root model [28]. The Yurke model accurately fits the behavior at longer separations but disagrees with the data at middle ones. However, it usually accurately predicts the time of coalescence. The observation of Svenšek and Žumer that the defects move at different velocities due to the flow is difficult to verify experimentally, as drift and Brownian motion do not allow us to measure the absolute radial velocities.

In the experiments of Pargellis et al. [16], defect annihilation was studied in very thick (≈ 155µm thick) films. Their data compared poorly against the square root model, since in their experiments the separation decreased linearly with time.

![Figure 2.5](image)

Figure 2.5: Two examples of the different models fitted to the separation $s(t)$ of two defects. The red line represents the square root model, while the blue line represents the Yurke model.

Every calculated $s(t)$ may be plotted against all of the others with each set moved such that they all show a common separation at a certain time, as in Fig. 2.6. This plot may be created over a wide range of separations and is used to find the similarities and differences in the behavior of different annihilation events.
Figure 2.6: Separation as a function of time, $s(t)$, for all data sets, shifted in time to line up at separation $= 30 \, \mu\text{m}$.

Figure 2.7: Average velocity as a function of separation, $< v(s) >$, for all events, compared against the Yurke model, in red. The velocity for each event is calculated by smoothing the raw separation as a function of time and making linear fits around each desired separation. Error bars are the standard deviation of the mean in the average calculations. Note that all velocities are negative, as the defects are moving toward one another.

The relative defect velocity, $v \equiv \frac{\partial s}{\partial t}$, is calculated by smoothing the $s(t)$ data and taking the first derivative. These velocities are smoothed, and the average velocity, $< v(t) >$, is determined as a function of defect separation. Because the motion is stochastic, some defects end up at a certain separation more than once; for these, each velocity at that separation is used to determine the
average. The results are shown in Fig. 2.7.

The Yurke model makes a prediction of the relative velocity, shown in Equation 2.4. This model, plotted in Fig. 2.7, shows broad agreement with the experimental data, but predicts a larger approach velocity at small separations than experimentally observed.

We see from Fig. 2.6 that there is a lot of variability in the defect trajectories in different annihilation events. To quantify the influence of stochastic forces on the defect motion, we determine the mutual displacement fluctuations, $\delta s$. Once the trajectories are aligned at a particular separation, we determine the variance, \( < s - < s(t) > >^2 \), at each separation, where \( < s(t) > \) is linear with slope equal to the average velocity at that separation. The slope of the variance versus time plot is linear, and proportional to a sort of “diffusion coefficient”: a measure of the randomness of the defect motion around that separation. This “diffusion coefficient” is shown in Fig. 2.8.

![Figure 2.8: Mutual displacement fluctuations as a function of separation.](image)

The overall average deterministic motion of all defect pairs is shown in the plot of \( < v(s) > \), Fig. 2.7, but the stochastic motion of the defects is shown in Fig. 2.8. While the defects speed up as they near each other, the stochastic component of the defect motion increases as well. If the stochastic forces do become more important as the defects come closer together, this may explain why the square root and Yurke models provide inadequate fits at smaller separations.

This suggestion that there is some additional force that we are currently unaware of the
Figure 2.9: Average velocity multiplied by separation, \( <v(s)> \cdot s \), as a function of separation. The black curve is the experimental data, the blue curve is the Yurke model, and the red line is the square root model.

significance of is further encouraged by looking at the \( s \cdot v \) curves. In the square root model, the quantity \( s \cdot v \) is constant with changing separation. The Yurke model predicts that \( s \cdot v \) increases with decreasing separation. However, the experimental data show a clear decrease of \( s \cdot <v> \) with decreasing separation, the opposite trend of Yurke, as shown in Fig. 2.9.

Figure 2.10: Orientations of the +1 defect (black) and the −1 defect (red) during an annihilation event. Orientation is defined as the angle at which the template (in Multitrack, see 1.4) had the highest correlation value for the initial time step.

Another interesting result is that orientation of the +1 defect changes much more than the
Figure 2.11: Schematic representations of the initial relative orientations of a pair of aligned (a) and unaligned (b) annihilating defects. The pair at left would be expected to annihilate more quickly than the pair at right.

−1 defect, as shown in Fig. 2.10. It seems reasonable that the mutual orientation of the defects would affect the annihilation dynamics. For instance, if the directors nearest each other had the same orientation on the two defects, as in 2.11(a), annihilation would presumably proceed more quickly than if they were not initially lined up, as in 2.11(b). This proposal is the subject of ongoing investigation.

2.4 Conclusion

After analyzing many defect pair annihilation events, we observe adequate fits to available $s(t)$ models at large separations, but poor fits at small separations. This is partially explained by an observed relative increase in stochastic effects at small separations. Theories of mutual separation velocity are also found incomplete and predict velocities that change more quickly with decreasing separation than experimentally observed. The observations here are significantly different than those obtained with thick films, suggesting that film viscosity or the surrounding air affects the defect dynamics more than current theories calculate.
Chapter 3

Effects of Surrounding Air Pressure on Single Defect Diffusion

We examine the diffusion of a single +1 defect within a smectic-C film at decreasing atmospheric pressures. Single +1 defects are induced in uniform two-layer films and in islands. Island motion may be modeled as that of an object in a 2D fluid surrounded by a 3D viscous material [5, 14]. Defects exhibit 2D Brownian motion, but this motion is affected by the drag of the 3D viscous medium surrounding it, i.e., the air.

3.1 Experiment

Freely-suspended films of smectic-C material are spread across a 3.35 mm diameter hole in a glass cover slide. The film is placed within a vacuum chamber, connected to a vacuum tank and pump, as shown in Fig. 3.1, a system designed to minimize both vibrations and excess air movement around the film. To obtain pressures from atmospheric (633 torr) down to about 50 torr, the chamber is opened to the vacuum tank, which is initially at very low pressure. For lower pressures, both tank and chamber are opened to the pump. Pressures are held steady to within 0.05 torr during the course of each experiment. The films are observed at room temperature via reflected polarized light microscopy, captured at a video frame rate of 30 fps.

The particular liquid crystal used, racemic MX8068 (Displaytech), is a commercial mixture with the following phase diagram: Crystal $\rightarrow$ ($-22^\circ$C) $\rightarrow$ SmC $\rightarrow$ ($60.5^\circ$C) $\rightarrow$ SmA $\rightarrow$ ($78^\circ$C) $\rightarrow$ Nematic $\rightarrow$ ($80.5^\circ$C) $\rightarrow$ Isotropic. This material was chosen because it has a broad smectic-C phase at room temperature.
3.2 Theory and Previous Experiments

Islands and other types of inclusions move as if in a quasi-2D fluid and have a characteristic Saffman length dependent on the island thickness and the viscosities of the film and the surrounding medium, as discussed in Chapter 1.2. If defects moved like islands, as a finite-sized inclusion, they would have a bigger Saffman length than if they just moved as a result of reorientation of molecules. This Saffman length would change as a function of the air viscosity, which depends on the pressure of the surrounding air.

In earlier experiments in our laboratory, oil droplets were suspended in SmA films and their mobility was observed as a function of decreasing air pressure [33]. The results, summarized in Fig. 3.2, show that the mobility of such inclusions increases with decreasing pressure. The observed behavior is consistent with HPW theory at moderate pressures but at very low pressures the motion of the inclusion reflects its confinement in 2D. My experiment set out to determine whether SmC topological defects behave in a similar fashion.

3.3 Results

3.3.1 Mobility and Decreasing Pressure

We observed topological defects in two slightly different environments. In one set of experiments, we measured the diffusion coefficient of a single defect in a two-layer film the size of the
film holder. The film starts at atmospheric pressure. The diffusion of the defect is then observed at various pressures, decreasing from atmospheric to 0.4 torr. This is not the minimum pressure of the vacuum setup, which is $\sim 10^{-3}$ torr, but rather the lowest pressure used in our experiments, for reasons explained below. The position of the defect is determined by using MultiTrack, described in Chapter 1.4. In the second set of experiments, a defect was observed within an island that was itself diffusing within the two-layer film. The island and defect are tracked using MultiTrack, and the island center position subtracted from the defect position to get the relative motion of the defect with respect to the island center. The diffusion coefficients of the defects are found using the Python program of Nguyen described in Chapter 1.4. A summary of the results is shown in Fig. 3.3.

We observe that the mobility of the defects increases as the surrounding air pressure decreases. The similarity of these results to the oil droplet experiments suggests that defects move as inclusions do: as finite-sized particles. However, in SmC films, when the fluid moves, the molecules both move and reorient, as opposed to in SmA films where they simply move. This implies another possibility for the results seen here: that the elastic constant, which determines how “springy” the film is, changes as a function of pressure.

To determine how or if the elastic constant changes as a function of pressure, we look at a 2D Boltzmann probability distribution. The defect may be considered to be confined to a potential
Figure 3.3: Diffusion coefficient vs. air pressure (log scale) for a defect (a) “free” within a two-layer film (diameter = 3.35 mm), and (b) within an island (diameter = 90 µm). A simplified diagram of the geometry is in the upper right of each plot.

well defined by the elasticity and boundary conditions of the film [11]. This is modeled by

\[ U = \frac{1}{2} Cr^2, \]  

(3.1)

where \( U \) is the potential, \( C \) is the spring constant, and \( r \) is the distance from the film (or island) center. This spring constant is related to the Frank elastic constant, \( K \), of the film by

\[ C = \frac{2\pi K}{R^2}, \]  

(3.2)

where \( R \) is the radius of the film. Therefore, to estimate the elastic constant of the film at each pressure, we calculate the number of instances the defect was at a certain distance from the center (of the film or island, depending on the system) and take that as the 2D Boltzmann probability distribution, \( P(r) \). Then, we solve for \( K \) given the equations above, as

\[ \int 2\pi r P(r) dr = 1 \]  

(3.3)

\[ P(r) = e^{-\frac{U(r)}{k_B T}} \]  

(3.4)
The slope of (b) is proportional to $C$.

Figure 3.5: Calculated elastic constant of the film as a function of pressure, for a defect in an island.

\[
\ln(P) = -\frac{Cr^2}{2k_BT} \quad (3.5)
\]

\[
\frac{\ln(P(r))}{r^2} = -\frac{C}{2k_BT}. \quad (3.6)
\]

Since the slope of $\ln(P)$ with respect to $r^2$ is proportional to $C$, it may be measured to obtain $C$ and $K$.

The elastic constants found this way, shown in Fig. 3.5, do not show a significant trend as a function of pressure. This data therefore implies that the change in mobility seen in Fig. 3.3 are not due to a change in elastic constant and that defects move as a core of several molecules diffusing together, like inclusions.
3.3.2 Defect Disappearance

The lower limit on the pressure used in the experiments is not due to limitations of the vacuum pump but rather to the disappearance of the defect after about five minutes at around 0.4 torr. As the pressure is reduced from atmospheric, the contrast between the brushes of the defect decreases until it disappears entirely. This is not unique to the defect itself; the $c$-director fluctuations in the film vanish as well and the film is characterized by an $n$-director parallel to the layer normal. This is consistent with having induced a SmC-to-SmA phase transition. This is interesting because while racemic MX8068 has a SmC-to-SmA transition, this occurs at around 60.5°C, which is far above the temperature ($\sim 22$°C) at which the film experiments are conducted.

There appear to be no previous reports of a SmC-to-SmA phase transition induced by lowering the surrounding air pressure, although the opposite effect has been observed at very high pressure ($\sim 100$ MPa) [30].

3.4 Conclusion

We observe the diffusion of a $+1$ defect as a function of the surrounding air pressure, both within an island and in a uniform, two-layer film. The mobility of the defect increases with decreasing pressure in a similar manner to inclusions in a SmA film. Since no significant change in elastic constant as a function of pressure is found, this implies that defects move as discrete group of molecules, rather than as a local reorientation of the molecules. An apparent SmC-to-SmA phase transition is observed for the first time at low pressure.

Further work will include a more in-depth characterization of the phase transition, including a direct measurement of tilt angle as a function of pressure to rule out phase separation as a contributing factor. The entire experiment will also be repeated using a pure material instead of a mixture to observe defect behavior at even lower pressures in an effort to see direct 2D confinement as seen for inclusions.
Bibliography


[33] Zhiyuan Qi. 2D brownian motion of inclusions in smectic a liquid crystal film (personal communication).