Hydrodynamics of smectic liquid crystal films

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Hydrodynamics of smectic liquid crystal films

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Eric Minor

Hydrodynamics of smectic liquid crystal films.

Thesis guided by Joseph MacLennan

Smectic A and C liquid crystals are capable of forming incredibly thin films, discretized by the number of molecular layers. This property makes liquid crystal films ideal for studying 2D hydrodynamics, a field of great interest both due to its fundamental importance to physics and because of its applications to biological systems. The phospholipid bilayer which makes up cell membranes is itself an ideal 2D fluid as it consists of only two layers of phospholipids, however cell membranes are incredibly small and difficult to work with. Liquid crystal films can be several millimeters across and stable for long periods of time. As part of the OASIS NASA experiment, a large number of experiments on bubbles of liquid crystal film in zero gravity were carried out. This thesis will use video of those experiments to draw conclusions about the hydrodynamics of smectic liquid crystal films without the influence of gravity. Analysis was performed of thermomigration experiments, coarsening experiments, and on terrestrial smectic C island diffusion experiments. A simulation based on the current understanding of coarsening, diffusion, and Ostwald ripening was constructed and compared to the experimental data, confirming our theoretical expectations. For the thermomigration experiments, thermally induced motion was observed and measured. The data suggest that the Marangoni effect is primarily responsible for this motion. For the smectic C analysis, where smectic C islands tended to form chains, it was found that islands in the chains tended to diffuse more in a direction perpendicular to the chain than in a direction parallel to the chain.
Chapter 1: Introduction

2D fluids form the basis of many important biological systems, however the most accessible mean for studying the hydrodynamics that govern such systems is using liquid crystal films. In biological systems, the phospholipid bilayer of the cell membrane forms an ideal 2D fluid [16] that is two molecular layers thick. Embedded in the membrane are proteins, which diffuse via Brownian motion. A direct study of membrane hydrodynamics is incredibly difficult due to the incredibly small size of the membrane. Liquid crystal films [17] are also able to form thin films, often only a few molecular layers thick, and several millimeters across. Liquid crystal films also contain various features that make the task of observing underlying hydrodynamics much easier. Namely, liquid crystals can contain features known as islands, which are thicker regions of film that act as if they are cylinders embedding in the film itself. This largely eliminates the need for extraneous objects to be embedded in the film for the purpose of observing flow. These islands experience Brownian motion, thermally induced motion, and Ostwald ripening. In addition, smectic C liquid crystals can contain topological defects which occur in pairs and have the appearance of crosses when viewed under crossed polarizers. The alignment of these defects can have significant impact upon their diffusive properties. This thesis is primarily concerned with studying the hydrodynamics of such features.

Liquid Crystals

Matter exists in any of several physical states, which correspond to various intermolecular degrees of order. Commonly known are the solid, liquid, and gaseous phases of matter. Solid matter has a very high degree of order. Molecules or atoms typically maintain a fixed distance from each other and follow a regular ordering pattern. Gaseous matter maintains almost no order and maintains no set distance. Molecules move about at random and, if no containing force is present, will move far away from one another. Liquid molecules maintain a relatively fixed distance from one another but have very little order. The molecules have no set pattern and move around essentially at random, although they will not move indefinitely far away from one another like a gas. Liquid crystals are phases of matter [1] between solid and liquid. Many different liquid crystal phases exist, each with a different types of order. Of primary interest to this thesis are the smectic A and smectic C liquid crystal states. The molecules discussed in this thesis can be thought of as rod-shaped [2] in both phases. In both smectic A and smectic C phases the molecules arrange themselves into layers, with each layer being a single molecule thick. The rod-shaped molecules align along a particular vector called the director. In smectic A films, the director primarily points perpendicular to the plane of the film. Smectic C films have a tilted director, allowing the molecules to point in any direction on a tilt cone. The molecules are still locally aligned, but the director can gradually reorient along the film. The states of matter also generally correspond to different thermal energies. Solid matter has the lowest energy, followed by liquid and then gas, which possess the highest amount of thermal energy. Correspondingly, liquid crystals have thermal energy between that of a liquid and a solid, with smectic C possessing less thermal energy than smectic A.
Thin films of smectic A or C phase liquid crystal can be primarily created in two forms. Flat films are made by drawing liquid crystal material over a small aperture with a spreader. Despite the fluidic ability of liquid crystals to spread, the LC material possess strong intermolecular attraction that prevents the film from breaking while being spread. Although the film may appear qualitatively similar to a soap film, there are important differences. A soap film consists of water trapped between two layers soap, while a liquid crystal film consists only of liquid crystal layers. Each layer is only a few nanometers thick, and films can be created consisting of as few as two layers. Soap films also pop after a short time due to the contained water evaporating. Although liquid crystal films undergo some evaporation over time, the rate is incredibly small compared to water, which allows the films to remain stable for extended periods of time [24]. Flat films form a meniscus at boundary between the film and the holder. The meniscus is much thicker than the film, which makes observations near it undesirable for the purpose of understanding the dynamics of 2D fluids. The thicker the region of film being observed, the more the hydrodynamics tend to become three dimensional.

The second method for obtaining a thin liquid crystal film involves creating a bubble instead of a flat film. To generate the bubble, liquid crystal material must first be inserted into a hollow needle. By gently pumping air through this hollow needle, a bubble can form and expand made out of the liquid crystal material. Similar to the flat film, the surface of the bubble is made of a few liquid crystal layers. Aside from a slight curvature, the bubble and the flat film are similar in thickness and hydrodynamic properties. The primary advantage of making a bubble instead of a flat film lies in the minimization of the meniscus. The meniscus forms at the boundary between a film and the object that film is tethered to. For the bubble, this boundary
consists only of the very small region where the inflation needle meets the bubble, as opposed to the flat film where the boundary surrounds the entire film.

The three materials referenced in this thesis are:

SN001: Polar SmA (mixture of 8CB + Displaytech MX 12160), Isotropic 56 °C Nematic 54 °C SmA 5 °C Crystal

SN003: Chiral SmC* (Displaytech MX 12805), Isotropic 84.7–82.0 °C Cholesteric 81.4 °C SmA* 66.1 °C SmC

SN004: Non-Polar SmA (Displaytech MX 12160), Isotropic 51.1 °C SmA -3.2 °C Crystal (3.1 °C SmA)

MX8068 (Displaytech): Isotropic 80.5 °C Nematic 78 °C SmA 60.5 °C SmC -22 °C Crystal

In the experiments, SN001 and SN004 are both kept in the SmA phase while SN003 and MX8068 are kept in the SmC phase.

Islands

On the surface of a liquid crystal film, regions containing excess material may exist. Where excess material exists, surface tension forces that material into a circular shape. These regions are known as islands. Like the bulk film, the liquid crystal material that comprises islands forms itself into discrete layers. An island may be as thin as a single layer thicker than the background film, or it may be tens of layers thicker. Although various effects, which shall be discussed later, can change the radius of an island, they are generally stable in shape and size. Islands generally appear brighter than the background film due to interference. Thicker islands will begin to take on a variety of colors.

A variety of methods exist for generating islands. One of the simplest and most effective involves blowing air over the surface of the film, engendering turbulent flow that pulls excess material out of the meniscus. This excess material is then forced to form islands on the surface of the film. Some islands naturally form from excess material that existed when the film was created. Islands can also form around nucleation particles, such as dust or ash. Phase transitions between liquid crystal phases can also generate islands. Care must be taken when using any of these methods, since too large of a disturbance can easily pop the thin film.

Islands can be treated as cylinders embedded in the background film, and they undergo Brownian motion similar to the motion of small particles in a fluid.
OASIS

The thermomigration, Ostwald ripening, and coarsening experiments were all performed on smectic bubbles onboard the International Space Station as part of the OASIS mission [8], in collaboration with NASA.

FIG 2: OASIS bubble chamber. The chamber contains the bubble inflation needle, airjet needles, camera lenses, and an ink jet droplet device. The ink jet is not used in our experiments.

Performing the experiments in microgravity mitigated or eliminated several factors that impede ideal 2D fluid mechanics observations on Earth. For the thermomigration experiments, microgravity eliminated the effects of buoyancy. This means that convection would be eliminated. Microgravity also prevents excess material from pooling around the bottom of the bubble (sedimentation). Islands are affected by gravity, and, in the presence of gravity, will move towards the lowest point in a bubble. This can cause islands to cluster in one region and prevent other interesting phenomenon from being observed.
The OASIS setup also contained airjet needles situated near the top of the bubble. Creating a stream of air from these needles creates rapid, turbulent flow on the surface of the bubble. Doing so generates an island emulsion on the surface of the bubble. Heating elements are wrapped around two of the airjet needles, which allow the bubble to be heated from above. The inflation needle is perpendicular to the airjet needles and is used to inflate the bubble. Liquid crystal material is injected into the needle, which then has air blown through it to inflate the bubble, which typically has a radius of approximately 8.2 millimeters. The inflation needle also contains heating elements, allowing the bubble to be heated from below. Both the airjet needle heaters and the inflation needle heater supply heat to specific locations, making them useful for creating temperature gradients along the bubble’s surface. The chamber temperature is controlled by heaters in the chamber walls.

Video was recorded using two Prosilica GX 1050C cameras, which each offered 1024x1024 resolution. One of the cameras, referred to as the micro view camera, was oriented to look at the bubble from a position opposite the inflation needle. The micro view camera was attached to optics that yielded a field of view of approximately 500 μm x 500 μm. The second camera, which will be referred to as the macro view camera, was positioned perpendicular to the inflation needle. The attached optics gave the macro view camera field of view of 18 mm by 18 mm. The micro view camera allowed for detailed measurements to be made of individual islands, while the macroview camera allowed measurements to be made of the island emulsion as a whole.

FIG 3: The inflation and airjet needles. Material is injected into the inflation needle, and a stream of air is used to generate the bubble. Airjet needles located near the top of the bubble can create rapid flow on the bubble surface to generate an emulsion of islands. Heating elements on two of the airjet needles and on the inflation needle are used to create a temperature gradient.
FIG 4: Islands moving on a bubble due to the application of the airjet needs. The airjets are used to rapidly rotate the liquid crystal material comprising the bubble. When the airjets are turned off, an emulsion containing a large number of islands remains. Also visible, the heater wire wrapped around one airjet needle.
Chapter 2: Coarsening

Coarsening in these is the gradual increase in the average radius of an emulsion of islands on a smectic liquid crystal films. This phenomenon was observed occurring in several bubbles of SN001 material where island emulsions were created using the airjets and then allowed to evolve over time without interference. The standing hypothesis is that coarsening mainly occurs via two main mechanisms, Ostwald ripening and coalescence.

Ostwald Ripening

Ostwald ripening is a process that was originally observed in supersaturated solutions, and was first described in 1896 by Wilhelm Ostwald. In supersaturated solutions [3], solid material exists in equilibrium with a saturated solution. Particles from the solid dissolve into solution at the same rate particles from solution deposit on the solid. In many cases, the concentration of the solution necessary to obtain equilibrium with the solid varies depending on the shape of the solid. Small condensates often require a higher solute concentrate to remain in equilibrium than larger condensates due to the change in vapor pressure at a phase interface [25]. Because of this, supersaturated solutions containing both large solid particles and small solid particles will attain a solute concentration somewhere between the equilibrium concentration for the small and large particles. The small particles will thus lose mass to solution faster than new mass deposits, while the large particles will have new mass deposited at a higher rate than the rate at which the large particle losses mass to solution [18]. It will appear that, in a supersaturated solution containing both large and small particles, large particles will gain mass while smaller particles lose mass.

Although a qualitatively similar process occurs in liquid crystal films, which we still tentatively refer to as Ostwald ripening, the exact mechanism is still the subject of research. Instead of particles in a supersaturated solution, we have islands in a liquid crystal film. The concept of solute concentration does not directly apply to islands and liquid crystal films, since there is neither a solvent nor a solute, only the liquid crystal material. Still, it has been observed that small islands tend to lose mass while larger islands gain mass. The most probable explanation is that the background film maintains a pressure somewhere between the equilibrium pressure of small islands and large islands. Similar to condensates in solution, islands are capable of both absorbing and relinquishing molecules from the background film. The rate at which these processes occur is governed by pressure instead of by concentration. Smaller islands require a larger background film to maintain equilibrium than larger islands. Thus, in a similar manner to precipitates in super saturated solutions, small islands lose mass while larger islands gain mass. Because the system is thought to be governed by pressure instead of concentration the associated rate laws are not well understood, nor the exact circumstances that must exist for Ostwald ripening to occur.
Observations of Ostwald Ripening

Fig 5: Two pairs of islands undergoing Ostwald ripening. The small islands shrink without ever making direct contact with the boundaries of the larger islands, which is indicative of Ostwald ripening. The larger islands are observed to grow.

Using video from the micro view camera of islands made from SN001, the behavior of individual islands or pairs of islands was observed. An effect similar to Ostwald ripening occurred when a small island moved close to a large island: the large island had its area increase while the small island experienced a corresponding area decrease. Summing the area of both islands in a pair reveals that the combined area of the pair remains fairly constant, despite the area of the two separate islands changing. This suggests that the lost mass of the small island is being transferred to the larger island, rather than ending up in the film, which would increase overall bubble size, or the meniscus.

Fig 6: A pair of islands undergoing Ostwald ripening. The large island grew while the small island shrunk. The sum of island areas remained constant, suggesting an effectively direct transfer of mass from the small island to the large island.
Observations were also made of small islands whose area did not begin to change significantly until they moved closer to larger islands, at which point the small island began to shrink while the larger island grew. This supports the idea that a short-range interaction is being observed. This appears to be different from Ostwald ripening in supersaturated solutions, which does not predict pairwise interactions, but instead predicts long distance interactions between all precipitates in a solution via interactions with the bulk fluid. An observation was also made of a small, bright island gaining mass when paired with a larger, dim island. As previously mentioned, island brightness is an indicator of greater thickness due to interference. This suggests that the island’s mass, and not overall boundary length, is the primary determining factor for which island gains mass and which loses mass.

Fig 7: An island undergoing Ostwald ripening and a plot of its change in area over time. Island only grows after moving closer to large Island. This demonstrates that in some cases, proximity between islands is necessary for mass transfer to occur. The small, bright island grows when it moves closer to the larger island. This suggests that the total mass of the island, which depends on both island radius and on island brightness, is more important than just the island radius.

Instances of small islands losing mass without any nearby islands was also observed. This phenomenon is more similar to traditional Ostwald ripening than the previously discussed short-range interactions. According to the Kelvin equation, the pressure necessary for an island to maintain equilibrium with the surrounding bulk fluid is dependent on the size of the island. Large islands require a lower pressure to maintain equilibrium than small islands. Thus, in an emulsion containing both small and large islands, the background has a pressure between the equilibrium pressures for small and large islands. Islands with equilibrium pressures similar to the pressure of the bulk experience no change in area. Large islands with equilibrium pressures much smaller than the bulk pressure will slowly gain mass from the bulk, while small islands with equilibrium pressures higher than the bulk pressure will slowly lose mass to the bulk. The change in island size via Ostwald ripening occurs slowly, taking around 23 minutes in some cases for a small island to completely disappear. Other islands shrink much faster and can disappear in as few as two minutes.
Fig 8: A small island that shrinks without nearby islands. The small island moves across the frame from left to right while shrinking, which suggests that proximity to more massive islands is not always necessary for mass loss to occur. This is similar to traditional Ostwald ripening where each island mainly interacts with the background fluid.

Ostwald Ripening of Holes

Holes are regions in liquid crystal films where the number of layers present is less than in the background film. While islands typically appear brighter than the background film, holes appear darker. Holes have been previously observed [23] in liquid crystal films, however the OASIS experiments were the first instance of an emulsion of holes. Ostwald ripening was observed occurring for several holes present on a bubble of SN003 material. Although a clear video of a hole growing due to Ostwald ripening was not available, observations through the macro view lens showed that the average radius of holes in the film increased over time.
Fig 10: Bubble containing and emulsion of holes and the Average hole radius over time for a bubble of SN003. The plot shows the data (blue dots) and fitting curve (red dotted line). The average hole radius increased over time, suggesting that coarsening also occurs for emulsions of holes.

Fig 9: An emulsion of holes and islands. Although the smectic C material used in this experiment inhibits coalescence events, Ostwald ripening is still able to occur.
Coalescence and Brownian Motion

Coalescence is the merging of two islands that occurs when the boundaries of those islands collide. When this occurs, the two islands quickly merge together into a larger, circular island with the combined mass of both of the smaller islands. The actual merging process occurs on a much shorter time scale than the time scale over which coarsening occurs, meaning that the change in the average island radius due to coalescence is primarily limited by the rate at which collisions occur [9]. The collision rate is governed, in turn, by Brownian motion [11].

Although liquid crystals are not three-dimensional fluids, each layer allows for the fluidic motion of its which molecules which means that objects embedded in liquid crystal films undergo Brownian motion. First discovered in 1827, Brownian motion is the seemingly random movement of small objects in a fluid. On a molecular scale, this motion is caused by the random collisions of molecules with the visible objects, causing small, random shifts in momentum. Islands in a liquid crystal film undergo this same type of motion. As molecules in the surrounding fluid collide with the island, the island undergoes a random walk.

Einstein quantitatively described this phenomenon in 1905 [10]. The Einstein relation establishes a diffusion constant that dictates the rate at which a particle undergoes Brownian motion. This constant is not universal but is instead determined by situational factors. The
relation is given in equation (1), with \( \mu \) being the particle’s mobility, which is the ratio \( \mu = \frac{v_d}{F} \) of the particle’s terminal velocity and an applied force, \( k_B \) which is Boltzmann’s constant, and \( T \), the absolute temperature.

\[
D = \mu k_B T \quad (1)
\]

\( D \), the diffusion constant, is related to the square of the average displacement the particle experiences in a unit of time, which is colloquially referred to as the Mean Squared Displacement (MSD). For a time step of size \( t \) in one dimension, the MSD is given as

\[
< x^2 >= 2Dt \quad (2)
\]

However, liquid crystals are two dimensional fluids and we must thus adjust our definition to account for two dimensions instead of one. This can be done be simply combining two, one dimensional MSD equations.

\[
< x^2 >= 2Dt \\
< y^2 >= 2Dt \\
< r^2 >= < x^2 + y^2 > = 4Dt \quad (3)
\]

For the purposes of calculating mobility, islands are modeled as cylinders embedded in a 2D fluid, which is itself immersed in a 3D fluid. The 2D fluid is the liquid crystal film, and the 3D fluid is the surrounding air. The mathematics of this setup were derived by Saffman and Delbrück [12]. A characteristic length scale, the Saffman length, is given by

\[
l_s = \frac{n_{2D}}{\eta_{\text{surround}}} \quad (4)
\]

where \( n_{2D} \) is the 2D viscosity of the liquid crystal material and \( \eta_{\text{surround}} \) is the viscosity of the surrounding air. \( n_{2D} = h \ast \eta_{LC} \), where \( h \) is the film thickness and \( \eta_{LC} \) is the bulk viscosity of the liquid crystal. The film is surrounded on both sides by air, so \( \eta_{\text{surround}} = 2 \eta_{air} \). Using these terms in (4) yields

\[
l_s = \frac{h \eta_{LC}}{2 \eta_{air}} \quad (5)
\]

If an island has radius much smaller than \( l_s \) mobility takes the form [13] of

\[
\mu = \frac{1}{4\pi\eta_{LC}h} \left( \ln \left( \frac{l_s}{r} - \gamma \right) \right)
\]
Where $\gamma$ is Euler’s constant of 0.577 and $r$ is the particle radius.

If an island has a radius much larger than $l_0$ then the mobility [14] is

$$\mu = \frac{1}{16\eta_{air} r}$$

(7)

The dependency on the viscosity of the liquid crystal material has been completely mitigated, as the island is interacting more strongly with the surrounding air than it is with the thin film.

In practice, islands often occur with a radius that is on the same scale as $l_0$, making both equations fail to accurately describe the island mobility. In 2008, Petrov and Schwille published a paper describing an analytic solution to the problem. The equation makes use of the reduced radius, $\epsilon = \frac{r}{l_0}$ and four fitting parameters, $b_1 = 2.748$, $b_2 = 0.515$, $c_1 = 0.738$ and $c_2 = 0.522$.

$$\mu = \frac{1}{4\pi\eta_{LC} h} \left( \ln\left(\frac{2}{\epsilon}\right)^{-\gamma} + \frac{4\epsilon}{\pi} \left( \frac{c_1}{2} \right) \ln\left(\frac{2}{\epsilon}\right) \right) \left( 1 - \frac{c_1 e^{b_1}}{1 + c_2 e^{b_2}} \right)$$

(8)

This is the form [15] later used in simulation calculations, and is accurate in both 2D and 3D regimes.

Coarsening Simulation

In order to test whether our understanding of Ostwald ripening and coalescence fully account for macro-view observations of island radius evolution, a simulation was created and run that modeled both Ostwald ripening and diffusion.

The first stage of the simulation involves constructing an initial state similar to experimental data. To do so, a Matlab implementation of the Hough transform for circles was used to obtain the radius of all circles near the center of the bubble image (from the camera’s perspective). Only islands from the central region can effectively be measured this way, since
shape distortion begins to inhibit the performance of the circular Hough transform near the edges of the bubble. The detected radii were then sorted into 10 bins based on radii. All the islands in a bin were used to calculated the average radius of islands in that bin when the simulation conditions were generated. Although binning slightly decreases the similarity of the simulated starting conditions to the true starting conditions, doing so greatly improves the speed at which the initial conditions can be generated. The area in which islands were detected was taken to be representative of the distribution over the bubble as a whole. This yields the expected number of islands of each radius over the entirety of the bubble.

To create a valid initial setup for the simulation, we must place islands onto a sphere with the same distribution and density as the real-world experiment, while also making sure the islands do not overlap. To prevent overlap, a binary image representing the valid locations an island may be placed is maintained for each bin, with the x and y coordinates of the image mapping to theta (polar angle) and phi (azimuthal angle) coordinates on the sphere’s surface. Initially, each binary image consists only of 1’s, representing that all pixels are valid locations for islands to be placed. As each island is placed, the Haversine formula is used to calculate the geodesic distance between that island’s center and the other pixels in the binary image. A pixel is invalid for island placement if the geodesic distance between that pixel and the newly placed island is less than the radius of the newly placed island plus the radius associated with the bin which that particular binary image is associated with. When a pixel becomes invalid due to a new island placement, that pixel has its value flipped from 1 to 0, indicating an island of the associated bin radius cannot be placed there. Because all operations are reduced to operations on pixel coordinates and island coordinates, this process has been parallelized on the GPU to greatly accelerate the process.

Islands are placed in order of descending radius, since it is easier to fit smaller islands between larger islands than it is to fit large islands between small ones. Each time an island is placed, a pixel is chosen at random from all pixels set to 1 in the binary image that is associated with the radius of the island to be placed. When the island is placed, the pixels of each binary image are updated as previously described and the process was repeated until the number of placed islands for each bin radius equal the bin counts. This process ensures that the initial distribution of islands in the simulation matches the initial distribution in experiment, while also preventing overlapping islands.
FIG 12: The simulated bubble next to the experimental bubble. The distribution of islands over the surface of the simulated bubble closely matches the distribution of islands over the surface of the experimental bubble.

The next step is to simulate Brownian motion, coalescence, and Ostwald ripening over time. For the purposes of the simulation, the parameters of 8CB are used.

At 22 °C, 8CB has a viscosity of 0.052 Pa s [4], a bulk density of 0.96 g/cm [5], a thickness per layer of 3.17 nm [6]. The air has a viscosity of 1.827 x 10^{-5} Pa s [7]

Using equations (1), (3), and (8), the average distance an island moves over a small timestep can be calculated. (8) yields the island mobility, which can then be used in (1) to calculate the diffusion constant. Finally, (3) can be rewritten as

$$< r_{\text{rms}} > = 2\sqrt{Dt}$$

to give the average displacement in a time t. In accordance with traditional Brownian motion, the direction of the displacement is chosen at random. The reverse Haversine formula is then used to calculate each island’s new position on the sphere. The geodesic distance between all islands is then checked using the Haversine formula and if two islands have moved close enough to one another to have their boundaries touch, the islands are merged to form a larger island. The merged island has an area equivalent to the masses of the subsumed islands and a position that is the area weighted average of the original two islands’ positions. In experiments, the merging of two islands usually induces a slight movement in nearby islands, however this effect was judged to have a minor impact on coarsening rates and was ignored in the simulation.
Ostwald ripening in the simulations was given a rate similar to the rate observed in the Ostwald ripening experiments. Islands larger than the average radius had their area grow by approximately 4 μm² per second while small islands had their area shrink by 4 μm² per second.

The result of the simulation was a coarsening rate very similar to the experimentally observed rate. The simulation showed an average radius increase from 150 μm to approximately 164.4 μm while the experiment showed an average radius increase of 150 μm to 165.6 μm. Although the experimental data contains a large amount of noise due to inconsistencies in island detection, the shape of the produced curve, increasing with a gradually decreasing slope, is also similar. This suggests that the combination of Ostwald ripening and Brownian motion driven coalescence account for the majority of coarsening in the experiments.
Chapter 3: Thermomigration Experiments

Using the material designated SN004 (Displaytech MX 12160), which is a non-polar smectic A phase liquid crystal at room temperature, a series of tethered bubbles were inflated aboard the International Space Station. SN004 Material was injected into an inflation needle, which subsequently had air pumped through it, causing a bubble to form at the tip. All observed bubbles contained a wide array of islands of differing radius and thickness. The inflation needle was then heated to a temperature which varied by experiment. The chamber temperature was held constant at 26 °C. The application of heat caused the islands in all experiments to move away from the point of heating. This phenomenon was first documented in 1855 [22] and is traditionally known as the Marangoni effect or as the thermocapillary effect. The occurrence of the Marangoni effect in smectic liquid crystals is a field of active research [21].

FIG 14: Thermomigration of islands on smectic bubbles in microgravity heated (a) from below and (b) from above. In these experiments, the bubble blowing and airjet needles were respectively suddenly heated to $\Delta T=+14^\circ$C and $\Delta T=10^\circ$C above the ambient bubble chamber temperature of $T=26^\circ$C. The vectors show the average island velocities measured during the first two seconds after the temperature gradient was imposed. The average velocities around the equator are 14 mm/sec and 5 mm/sec respectively. Heating in (a) was achieved by heating the bubble blowing needle, to which the bubble is tethered at the southern pole. The bubble was heated from above (b) by raising the temperature of two of the airjet needles, one of which can be seen at the top of the images (the other is out of focus, behind the non-heated needle at left). In this example, the bubble was not centered below the airjet needles, so that the islands move down and to the left, towards the coolest part of the bubble. The liquid crystal material used here is MX 12805. The bubble diameters are approximately 15 mm.
Four experiments were conducted where the bubble was heated from below with the inflation needle, and one experiment was conducted where the bubble was heated from above by the airjet needles. Similar behavior was observed in both cases, however the bottom heating experiments will primarily be discussed due to the greater number of experiments performed. In order to determine the relative rates at which islands were induced to move by the thermocapillary effect, islands were tracked only for the first 30 seconds, during which the islands moved linearly. After the initial measurement period, secondary flows tended to form and the temperature gradient became less pronounced. Islands were tracked mainly using edge detection and morphological operations. Canny edge detection was used to find potential boundaries between islands and the background film. The edges were then morphologically thickened to fill in incomplete boundaries, and then thinned to make the boundaries only a single pixel thick. These boundaries were then used to define objects within the image. Most of this process was performed on the GPU, allowing it to complete very quickly (~0.2 seconds per image), however the program did falsely identify some video artifacts as islands. These objects had to be manually removed from the data.

FIG 15: Velocity vs polar angle of islands on a smectic bubble in a temperature gradient. In this experiment, the bubble chamber was first equilibrated at T=28 °C and then the bubble inflation needle was suddenly heated to a higher temperature T+ΔT, with ΔT equal to +22°C. The inflation needle (the “south pole” of the bubble) is at θ=0. The velocities were determined by tracking the motion of all islands in the field of view during the first two seconds after the needle was heated and computing their average speeds. The mean island velocity decreases linearly with polar angle (red lines).
The data showed that the velocity at which an island moved was directly related to how close the island was to the point of heating. In addition, the higher the temperature difference between the bubble and the needle, the faster the islands moved. The relationship between the temperature differential and both the slope and intercepts were also found to be linear.

FIG 16: Average velocity near the heated needle and velocity gradient on the bubble as a function of the temperature difference $\Delta T$ between the needle and the bubble chamber. The velocity gradients in (b) correspond to the slopes of the linear fits of island velocity with respect to angle shown in Figure 1. The velocity decays most rapidly when the temperature gradient is largest.

This provides strong evidence that thermocapillary motion is occurring. A competing explanation for why smectic islands moved when a temperature gradient was created suggested that the extra heat induced excess material to leave the inflation needle, and that the islands moved because there was an overall background flow. If this was the case, it would be expected that island velocities would drop near the equator and increase near the poles due to flow conservation. The linearity of the data instead supports the hypothesis that thermocapillary motion is occurring. Thermocapillary motion occurs solely due to a surface tension gradient induced by a thermal gradient and would not be affected by flow conservation. The thermal gradient is expected to decrease linearly with distance from the point of heating.
FIG 17: Velocity vs polar angle of islands on a smectic bubble in a temperature gradient compared with the behavior of a hypothetical incompressible fluid film originating at the south pole and disappearing at the north pole (blue dashed curve). The incompressible fluid flow line is adjusted to match the velocity measured at the equator. In this experiment, the bubble chamber was first equilibrated at $T=28 \, ^\circ C$ and then the bubble inflation needle was suddenly heated to a higher temperature $T+\Delta T$, with $\Delta T$ equal to $+22^\circ C$. The inflation needle (the “south pole” of the bubble) is at $\theta=0$. The velocities were determined by tracking the motion of all islands in the field of view during the first two seconds after the needle was heated and computing their average speeds. The mean island velocity decreases linearly with polar angle (red line).
Chapter 4: Smectic C Islands and Defect Diffusion

Experiments to determine the character of smectic C island and defect dynamics were performed in gravity, without association to the aforementioned OASIS project. The experiments were performed with a flat film using material MX8068.

As previously mentioned, smectic C liquid crystals have a tilted director. The director can change orientation throughout the film, causing interesting behavior due to the energy associated with changing director orientation.

FIG 18: diagram of director orientations around smectic defects, with red arrows representing director orientation. All islands contain a +1 interior defect and a -1 exterior defect. The director must be parallel to the island boundary, causing a +1 defect to exist inside all smectic C islands. A -1 exterior defect must also exist outside the islands.

The director is aligned with the boundary of islands. This causes a circular pattern to develop in the interior of islands, which also necessitates that a point somewhere inside the island where the director field has an undefined orientation [28]. This point is known as a +1 topological defect. The presence of a +1 defect inside each island also necessitates the existence of a -1 defect outside each island that balances the topological charge of the film.

An experimental analysis of films made from smectic C liquid crystal material was performed in order to better understand the diffusive characteristics of smectic C islands and their associated defects. The material used in the experiments was MX8068. In order to visually see the director orientations, polarized light was reflected from the film and passed through a
second polarizer. This caused the image to be black in regions where the director was either perpendicular or parallel to either of the polarizers.

![Image showing -1 and +1 defects](image)

**Fig 19:** An example image showing both the -1 and +1 defect associated with an island. The darker regions are created when incoming light is either polarized either parallel or perpendicular to the director orientation.

To analyze island and defect motion, two methods for extracting diffusion coefficients from motion data were utilized. The first method involves fitting a gaussian distribution to a histogram of island displacements over a small time lag. The second method plots the Mean Squared Displacement (MSD) of the tracked island over various time steps, and attempts to fit a straight line through the plot. The IDL software for performing this operation was developed by Joseph MacLennan at the University of Colorado Boulder.
FIG 20: The two methods used for determining the diffusion coefficient of an island. Both methods are 1 dimensional, with separate coefficients being calculated for x and y diffusion. Although both methods are effective, the MSD plot is generally chosen as it is easy to visually see the goodness of a produced fit.

Island Chains

Islands in smectic C films tend to form dipolar chains[29] [30], with the external -1 defects situated between the islands in the chain. The diffusion coefficients for the islands in the chain were calculated, both in the direction of the chain and perpendicular to the chain. The result showed that islands in the chain have higher diffusivity and mobility perpendicular to the chain as opposed to along the direction of the chain. However, the ratio of perpendicular to parallel mobility was not the same for all islands. Small islands in particular had a much greater anisotropy in their diffusion characteristics, while large islands only slightly favored diffusion in the perpendicular direction.
FIG 21: An example of islands in a chain. Although not visible here, -1 defects are present between the islands.

In figure 21, Island 0 had an x (parallel) diffusion coefficient of 0.0526 +/- 0.0029 \( \mu m^2/sec \) and a y (perpendicular) diffusion coefficient of 0.1598 +/- 0.0054 \( \mu m^2/sec \). Island 1 had an x diffusion coefficient of 0.0552 +/- 0.0032 \( \mu m^2/sec \) and a y diffusion coefficient of 0.1475 +/- 0.0101 \( \mu m^2/sec \). Both islands are similar in size and had perpendicular diffusion coefficients approximately 3 times as large as their parallel diffusion coefficients.
FIG 22: A longer island chain containing islands of various sizes. By analyzing the diffusive motion of islands present in this chain, we were able to ascertain the degree to which island size affects the ratio of perpendicular to parallel diffusivity.

A longer chain, shown in figure 22, was also found and tracked. The wide variety of island radii present in this chain allowed the relationship between island radius and diffusive anisotropy to be determined.
FIG 23: Plot showing the relationship between island radius and the ratio between parallel and perpendicular diffusion coefficients. A clear connection exists between island radius and the diffusion constant ratio.

The observed relationship, plotted above in FIG 23, shows a connection between island radius and the ratio of perpendicular diffusive motion to parallel diffusive motion. A preliminary explanation is the greater inertia of large islands causes them to be less influenced by the forces created by director orientation that cause small islands to be confined along the parallel dimension.

In order to confirm that islands were actually bound in a chain, and not merely arranged in a chain formation by happenstance, the Pearson correlation coefficients [20] between islands were calculated. The Pearson correlation coefficient is a measure of the degree to which two random variables are related. A 0 represents two random variables that have no connection, while a 1 represents two random variables that are perfectly correlated. The Pearson correlation coefficient is defined as the covariance of the two random variables divided by the product the two random variables’ standard deviations. The result of the analysis shows that motion along the chain axis for each island is highly correlated to the motion of the adjacent islands and correlated to a lesser degree with the islands further away.
FIG 24: Graph depicting how strongly correlated the motion of each island is with each other island in the chain. The line for each island peaks when correlated with itself (always one) and decreases the further away you move on the chain.

The relative strong Pearson correlation coefficients confirm that the islands are forced to move together as a chain. This result also shows how the correlation between the motion of islands in a chain decreases the further away two islands are in the chain.

Mutual Mobility

Measurements were also made of the mutual mobility of pairs of islands and between an island and its external -1 defect. The code for calculating mutual mobility was provided by Zhiyuan Qi. Single object mobility refers to the ratio between an object’s terminal velocity and an applied force. Mutual mobility [19] is the ratio between the terminal velocity of one object in response to the force applied to a second object. The mutual mobility is calculated and used as a means of measuring the tendency of one object’s motion to cause motion in a second object and vice versa, for a pair of objects.

Mutual mobility was first calculated for pairs of islands. The results showed that radial mutual mobility was higher than tangential mutual mobility for islands with a -1 defect between them. When islands are further apart, both tangential and radial mutual mobility components were lower. When no -1 defect existed between the island pair, with the islands and defects instead assuming a quadrupole [27] formation, radial and tangential mobility of the islands were much closer in magnitude.
FIG 25: Island pairs for which mutual mobility was measured. Both the separation of islands and the alignment of defects had significant impact on the observed mutual mobilities. The two islands in (a) had a defect directly between them and experienced a radial mutual mobility ($M_{rr}$) of 1.13 μm$^2$/s and a tangential mutual mobility ($M_{\theta\theta}$) of 0.38 μm$^2$/s. The two islands in (b) had a significantly larger separation and had a $M_{rr}$ of 0.53 μm$^2$/s and a $M_{\theta\theta}$ of 0.38 μm$^2$/s. The two islands in (c) had a small separation but had the defects aligned in a quadrupole, which yielded an $M_{rr}$ of 1.67 μm$^2$/s and a $M_{\theta\theta}$ of 1.47 μm$^2$/s.

The diffusion characteristics of -1 defects was also examined. The locations of defects were tracked using IDL pattern matching software. Using the defect tracks, the same techniques that were used on islands to calculate diffusion were also used on defects. The diffusion of defects locked between two islands was tracked to determine whether the same asymmetrical diffusion characteristics that were present for islands locked in chains were observed with defects. The result showed that although a slight diffusive anisotropy was present, it was of a much lesser magnitude than what was observed for islands in chains. The diffusion constant perpendicular to the chain axis was approximately 3-7% greater than the parallel diffusion constant. Although significant, this is much smaller than the 20-70% discrepancy seen in island diffusion.
Defects between islands. $D_\perp$ refers to the component perpendicular to the chain axis while $D_\parallel$ refers to the component parallel to the chain axis. The defect in (a) had a $D_\perp$ of 1.70 μm/s and a $D_\parallel$ of 1.77 μm/s. The defect in (b) had a $D_\perp$ of 2.00 μm/s and a $D_\parallel$ of 2.15 μm/s. Although the diffusion constants are slightly anisotropic, the anisotropy is not as strong as for islands in a chain.

The final piece of analysis performed was to determine the mutual mobility of an island defect pair. The Brownian motion of topological defects in liquid crystal films has previously been observed and studied[26], although not in terms of mutual mobility. The experiment showed that, similar to island pairs, the defect island pair had a much higher radial mutual mobility than tangential mutual mobility. The result was not unexpected, as -1 defects remain close to their paired island but do not seem to have a preferred angular position.

Mutual Mobility

Radial: 0.96 um$^2$/s
Tangential: 0.69 um$^2$/s
Total: 1.64 um$^2$/s

The pair had a $M_{rr}$ of 0.96 μm$^2$/s and a $M_{\theta\theta}$ of 0.69 μm$^2$/s.
Chapter 5: Conclusions

This analysis has revealed several important characteristics of smectic liquid crystal films. Islands were observed to exhibit behavior similar to Ostwald ripening, although the exact rules which govern the interaction for islands are not fully understood. Holes, regions in smectic films that are thinner than the background film, were observed in grow in certain experiments, a phenomenon which has yet to be observed in gravity. A simulation was carried out that implemented both Brownian motion-driven coalescence and the experimentally found Ostwald ripening rate. The rate of coarsening in the simulation closely matched the experimental coarsening rate, which confirms our hypothesis that coalescence and Ostwald ripening are the main driving factors behind the coarsening of smectic liquid crystal films.

When a thermal gradient is created across a bubble of smectic A material, islands are induced to move from hotter regions to colder regions. The velocity of the islands decreased linearly with geodesic distance from the point of heating. This provides evidence that the motion was caused by changes in the surface tension caused by a surface temperature gradient, which provides a force to push islands along the surface of the bubble. This phenomenon is known as the Marangoni effect.

Finally, the diffusive motion of smectic C islands and defects was analyzed. These islands tend to form chains, with -1 defects present between the islands. The islands have higher diffusivities perpendicular to the chain axis than parallel to the chain axis, and the anisotropy was found to be dependent on island radius. The mutual mobility of island pairs was measured and found to be dependent on the positioning of defects between the islands. Radial mutual mobility was always higher than tangential mutual mobility. However, when a -1 defect was directly between the islands the degree of anisotropy was maximized. Mutual mobility was found to decrease with distance. The mutual mobility between an island and its associated defect was also measured, and the radial mutual mobility was found to be higher than the tangential mutual mobility.
References


