Hydroelastic Interactions Between Inclusions on Freely Suspended Smectic C Liquid Crystal Films

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Abstract

Interactions between inclusions (called islands) of extra layers in smectic liquid crystal films grow much more complex when observed in smectic C films. In a smectic C film, the c-director is nonvanishing, and is constrained by inclusions to specific orientations. These distortions result in the formation of a +1 virtual point defect inside the island and a -1 defect in the surrounding film. The presence of topological constraints of the c-director field at the boundaries of islands directly contribute to elastic interactions within the film and between islands. Theory describing these inclusions so far only includes these elastic interactions, but discrepancy between experimental and theoretical distributions lead to the conclusion that hydrodynamics may play a near-equal role in mediating island interactions in smectic C films. We attempt to observe and quantify island interactions in smectic C films in order to understand the roles played by elastic and hydrodynamic interactions between islands in these films.

1 Introduction

Smectic liquid crystals (LCs) may form thin films when drawn across a circular aperture around three millimeters in diameter [1].These films tend to be multiple layers thick, and can have as many as hundreds of layers, or as few as two layers. In films only a few layers thick, disk-like inclusions of extra layers may form, called islands. These islands are part of the film and are subject to the overall fluid mechanics of the film as well as the molecular interactions.

In the smectic C phase, molecules have tilt with respect to the layer normal, and this tilt can be represented by a two dimensional vector field embedded in the plane of the film. This field is called the c-director field and is useful in understanding energy constraints in each layer. Distortions of this field have elastic energy cost, and therefore islands undergo elastic interactions mediated through the c-director field. The presence of islands sets topological constraints in the film which force the c-director to align parallel to the island boundaries when close.[2] The orientation of the c-directors just inside and outside the island boundaries (oriented clockwise or counterclockwise) demonstrates the chirality of the island itself, each option a mirror image of the other. Due to the constraints at the island boundaries, singularities of the c-director field form in pairs inside the island and in the surrounding film. These are known as virtual point defects [2] and a +1 defect forms inside the island while a -1 defect forms in the surrounding film.

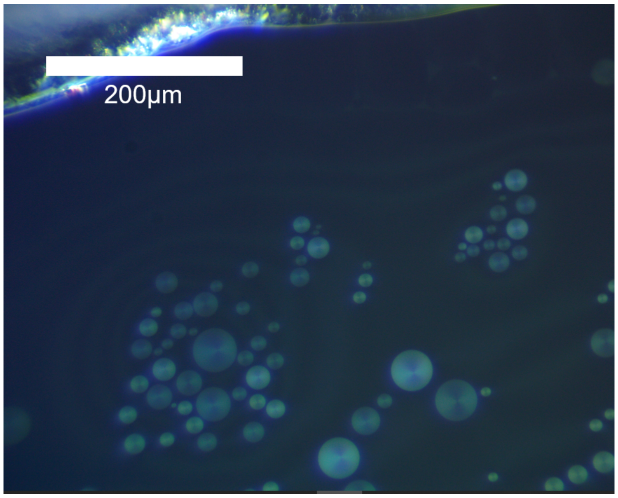

Figure 1: Smectic C islands shown under polarized light with slightly decrossed polarizers, using reflective microscopy. Two brushes are visible on each island signifying the +1 defect present at the center of each inclusion. The material shown is MX12846 which is racemic, meaning the islands can be either right-handed or left-handed

Equilibrium configurations of these defects naturally form in freely suspended smectic C films since the defects behave in the manner of signed point charges. In other words, islands in smectic C films have an equilibrium separation determined by the interactions between point defects. Quadrupolar configurations occur between two islands of opposite handedness (chirality) and longer dipole chains form between islands of the same handedness, in a pattern of alternating +1 and -1 defects. The motions of islands are in essence the motion of the film itself, and are therefore highly influenced by the c-director elastic interactions.

It has been shown that the hydrodynamics of smectic A fluid layers play a highly influential role in island interactions [3, 4], and these interactions are definitely present in the smectic C case, although much more complicated. When islands move, a surrounding flow field is created, affecting the motions of other islands. This is an expression of the viscous drag forces present, which would theoretically affect the c-director field as well, creating a sort of coupling between the two. This report contains preliminary experimental research into island pair diffusion.

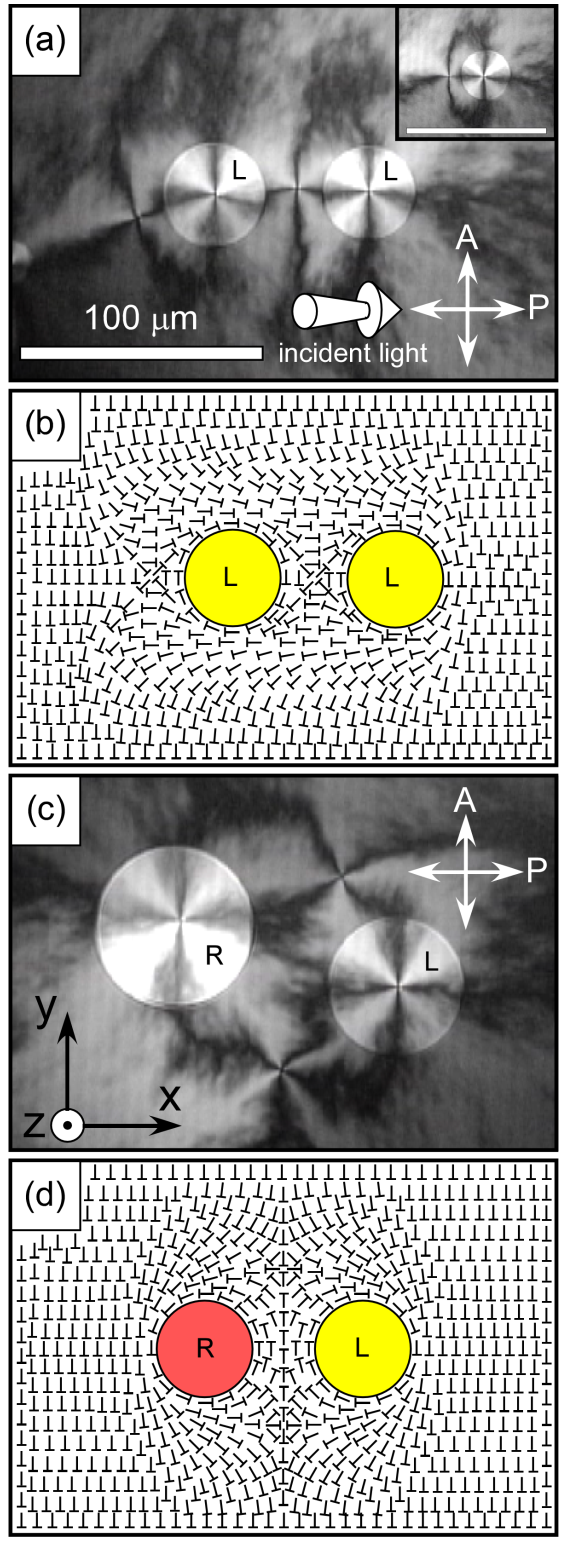

Figure 2: Equilibrium configurations of smectic C islands under polarized light, featuring a dipole (a) and a quadrupole (c). (b) and (d) are diagrams showing the director field orientation in the background film surrounding the islands. Island chirality is labeled inside each circle. Image taken from ref. [2].

2 Experimental Methods

This project is an experimental attempt to understand how elastic and hydrodynamic interactions combine in a two-dimensional medium. Freely suspended films, two layers thick, were created by spreading liquid crystal over a 5mm-diameter hole in a glass cover slip. In this experiment, we used Displaytech MX12846 and Displaytech MX12805, which are respectively racemic and chiral, and both of which are smectic C at room temperature. The viscosity of these materials is 0.06 Pa s [5] and the layer spacing can be assumed to be practically the same as 8CB, 3.17 nm [4]. The Saffman length can then be calculated using the equation $\ell_s = \eta h/(2\eta')$ [6] where η' is the viscosity of the surrounding fluid, in this case air, $\eta' = 1.827 \times 10^{-5}$ Pa s [4]. Thus the Saffman length for a two-layer film of each material is $\ell_s = 10.4$ μm, which I will use to create unitless values for the separation between islands.

Island pairs were observed using a microscope under 20x magnification, with slightly decrossed polarizers and using fluorescence microscopy. Videos of these pairs were captured using a Phantom high speed camera, at one hundred frames per second.

To find island centers and radii, machine learning techniques were implemented to analyze the frames of each video. Specifically, a support vector machine was used [7] and Canny's method of edge detection [8].

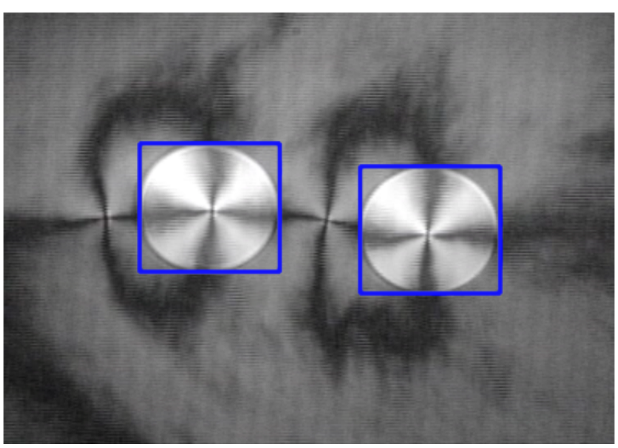

Figure 3: Example showing how the support vector machine find islands and marks them.

These methods provided the island centers and radii for each frame, allowing me to create trajectories for each island (as in figure 4). This allowed us to find the island separations in each frame, and the rest of the results in this report. Figure 4 gives a visual representation of how this process provides the island separations over time. Code was written to extract these values, and process them in accordance with the types of data analysis in this report.

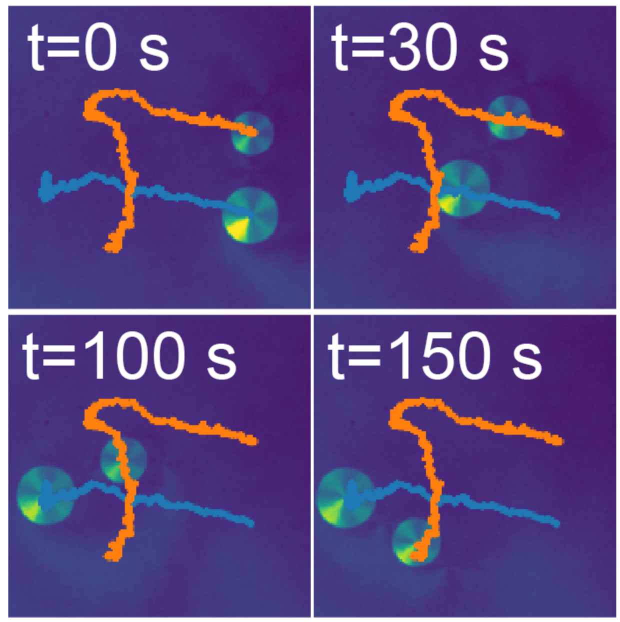

Figure 4: Trajectories of Islands in a smectic C film, the trajectories for each island can be seen (orange and blue) and the island positions at different times. The orange and blue plots represent the full trajectories of each island, and the islands are shown at different points along these trajectories. Islands in this figure are from a dipole video captured by Aaron Goldfain.

3 Experimental Results

3.1 Fluctuations in Separation

Due to the presence of an elastic potential on the film (since point defects behave as point charges), smectic C islands appear to exhibit an equilibrium separation between the island centers. Fluctuations around this separation can be measured and placed into a distribution to quantify the correlation between the island motions. Theoretical distributions have been generated based only on elastic theory.

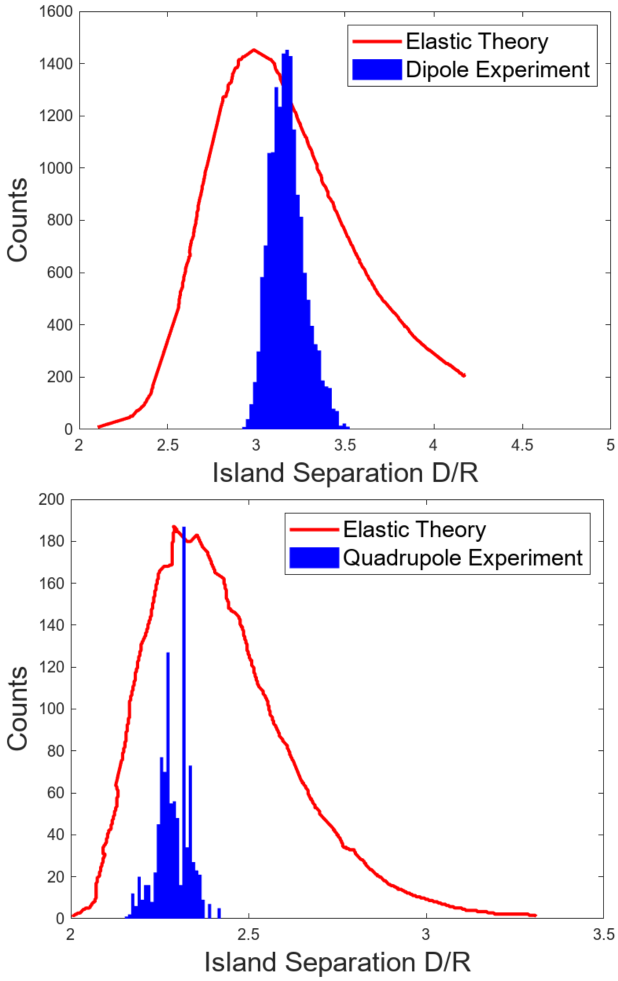

Figure 5: Distributions of island separation as predicted by elastic theory in both the dipolar (top) and quadrupolar (bottom) case. Island separation (D) is divided by island radius (R). Data collected by Aaron Goldfain.

The comparison between these models and the experimental results demonstrates the exciting possibility that the interactions between islands may be more complex and contain more types of interactions that previously thought. The resultant distributions are much narrower than theoretical predictions in both the quadrupolar and dipolar case. This could provide evidence for the possibility that the motions of the islands are more correlated than predicted by elastic theory, hinting that more types of interactions may be present.

As can be seen, both theory and experiment demonstrate an 'equilibrium separation', but the disagreement arises from the different results concerning the amount islands will fluctuate around the equilibrium separation. In both the dipolar and quadrupolar cases, elastic theory predicts much higher fluctuations than experimentally observed, providing evidence for other constraints that may be a factor. These distributions demonstrate that elastic theory alone cannot reliably model the interactions between islands in smectic C films, and that more complex physical interactions are taking place.

3.2 Mobility and Mutual Mobility

Diffusion constants and related mobilities can provide excellent insight into the nature of motion, especially in hydrodynamics. The single island mobility is a measure of the rate at which an island diffuses relative to a stationary background, essentially a measure of how much the island is "allowed" to move. The most conventional way to find this value is by computing the mean-squared displacement of the island (MSD). The MSD is computed by averaging the square of the displacement over different time intervals, and by plotting these values against the time interval, the diffusion constant can be extracted from the following equation.

$$\langle x^2 \rangle = 2Dt + V^2 t^2 \tag{1}$$

Where D represents our diffusion coefficient, V represents our drift velocity, and x is the displacement in that time interval. It has been shown that an object with drift in one direction may exhibit a parabolic MSD curve with this form. To find the mobility from these values, it is simply

$$\mu = \frac{D}{k_b T} \tag{2}$$

Where T represents the temperature (in these experimets, T is room temperature). Parabolic curves were shown for the MSD in both the quadrupolar and dipolar case (Figure x).

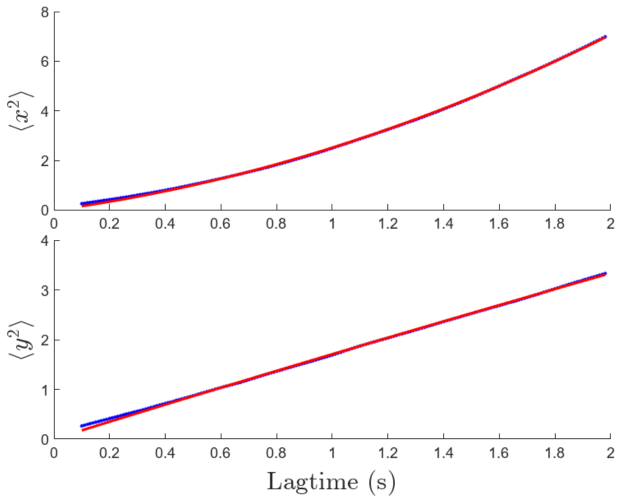

Figure 6: The mean squared displacement for a single smectic C island in a dipolar configuration, in the x and y direction. As can be seen, the y-direction appears almost perfectly linear, hinting that there was practically no drift in the y-direction. The experimenal data (blue, video by Aaron Goldfain) is shown with a parabolic fit (red). The x mobility found was 0.879 micron-squared per second.

The mobility is a good measure for the motion of a single island, but to observe how the motions of multiple islands are related, a new measure named the mutual mobility becomes useful. For the motions of two islands the mutual mobility tensor is shown in the following equation [3]

$$\mathbf{V}_1 = \mathbf{F}_{11}\mathbf{M}_{11} + \mathbf{F}_{12}\mathbf{M}_{12} \tag{3}$$

Where $\mathbf{M}_{12}$ is the mutual mobility tensor. When finding the components, it becomes useful to transform to a local coordinate system consisting of an axis parallel to a line connecting the island centers, and an axis perpendicular to the first one, drawn from the center of the first island. In these coordinates, the diagonals vanish and the only components remaining are the fully radial $\mathbf{M}_{12}^{rr}$ and tangential $\mathbf{M}_{12}^{\theta\theta}$, and these components can be treated as independent from one another (as they are related perpendicular axes). This allows us, as in the single mobility calculation, to find what is called the mutual diffusion coefficient in each direction with a very similar equation.

$$\langle r_1 r_2 \rangle = 2D_{rr}t + V^2t^2 \tag{4}$$

Where r_α represents the radial displacement of island α and D_{rr} represents the mutual diffusion coefficient in the radial direction. Plots in figure 7 are examples of these plots showing the parabolic fit and that provides coefficients. An equation identical to (4) can be used for the tangential direction as well.

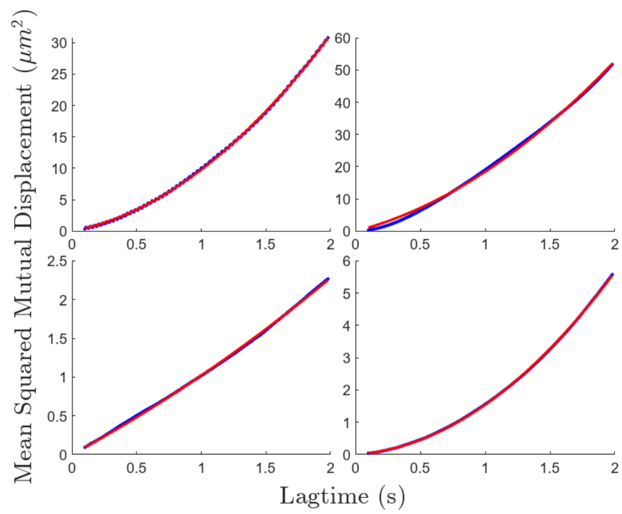

Figure 7: Mutual displacement versus lagtime shown for both the quadrupolar (top graphs) and dipolar (bottom graphs) case. Each data set produces two graphs, representing the radial (left) and tangential (right) directions. The parabolic fit (as in equation 4) is shown in red on top of the data points (blue). These fits can be used to extract the mutual mobility for each data set.

Exactly as in the single mobility calculations, the mutual mobility can be calculated from the mutual diffusion coefficient as shown:

$$\mathbf{M}_{12}^{\alpha\beta} = \frac{D_{\alpha\beta}}{k_bT} \tag{5}$$

For past experiments on smectic A films, theoretical predictions were demonstrated by modeling the mutual mobility of island pairs as a function of the average separation of said pairs. The comparison between this theory and smectic C data reveals the similarities and differences between smectic C and smectic A in order to understand which types of interactions may be occurring.

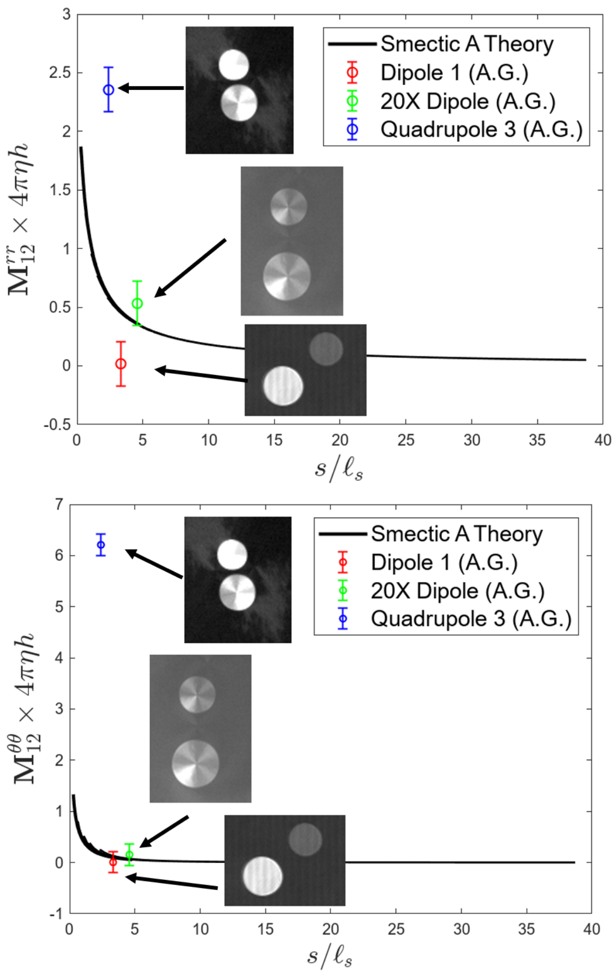

Figure 8: Mutual Mobility plotted versus the unitless separation of the island pairs, in the radial (top) and tangential (bottom) directions. Shown in comparison with smectic A theory, and in both the dipolar and quadrupolar case. The island separation is weighted with the saffman length. Images of each island pair are shown next to their data point and it can be seen that dipolar islands appear to fit with the theory better than quadrupolar islands, which seem to have much higher mutual mobility. The data sets used were collected by Aaron Goldfain (A.G.)

As in the figure, the dipolar data seems to be similar to the smectic A case, while the quadrupolar islands appear to have higher mutual mobility than smectic A islands. This would make sense if both hydrodynamic interactions and elastic interactions were present on smectic C films as the island motions would be more correlated.

4 Conclusion

In summary, the complexity of interactions in smectic C films lends itself to a more experimental approach. With previous studies on smectic A films revealing hydrodynamics as the central form of interaction, it is plausible to suggest that fluid flow mechanics may play a role in smectic C films as well. However, the prescence of elastic energy and interactions in these films makes the types of interactions difficult to discern and quantify, so multiple analyses were performed in an attempt to understand the nature of the mechanics on smectic C films.

Island pairs were observed in motion in two layer freely suspended smectic C films, and the resultant videos were analyzed for fluctuations, mobilities, mutual mobilities, in order to determine how the motions of the islands might be correlated or restricted. Mutual mobilities for dipolar islands were found to have similar trends to the smectic A hydrodynamic theory, whereas the quadrupolar islands seem to exhibit a higher mutual mobility, meaning the motions of the two islands were more correlated than expected of smectic A islands.

The key to combining hydrodynamics and elastic interactions in theoretical research is understanding how the the hydrodynamics and elastic interactions might complement each other. There is a layered coupling between the island motions and the elastic and hydrodynamic mechanics, but these mechanics are also coupled, making the theory much more complicated. It may be possible to simulate the coupling by showing how molecular interactions contribute to both fluid flow and elasticity, and expanding them to higher numbers. Experiments in smectic C films should continue to further observe exactly what shape the mutual mobility plots may take.

4.1 Acknowledgements

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