Observation And Analysis Of Smectic Islands In Space (Oasis)

Padetha Tin[#1], Noel Clark[*2], Joseph Maclennan[#2], Matthew Glazer[*2],
Cheol Park[*2], Ralf Stannarius[#3]

[#1]National Center for Space Exploration Research, NASA Glenn Research Center,
21000 Brookpark Road, Cleveland, OH 44135

[#2]Ferrolelectric Liquid Crustal Institute, Department of Physics
University of Colorado-Boulder, Boulder, CO 80309

[#3]Department of Physics, University of Magdeburg,
Magdeburg D-39106, Magdeburg, Germany

[1]padetha.tin@grc.nasa.gov

***Abstract*— The OASIS (Observation and Analysis of Smectic Islands In Space) space flight experiment is a series of experiment of the interfacial and hydrodynamic behavior of freely suspended liquid crystal (FSLC) films which are the thinnest known stable condensed phase structures making them ideal for studies of fluctuation and interface phenomena. FSLC films in microgravity present extraordinary opportunities for the study of fluid dynamic and thermodynamic behavior in reduced dimensionality, and for the exploration of fundamental nonequilibrium fluid interfacial phenomena. Detail tests of theories of hydrodynamic flow, of relaxation of hydrodynamic perturbations, and of hydrodynamic interactions in 2D will be done on freely suspended bubbles in microgravity, with and without islands/domains, ideally providing physically and chemically homogeneous 2D fluid systems for the precision study of 2D hydrodynamics. The proposed experiments will be carried out in dedicated sample chambers in the Microgravity Science Glovebox (MSG) of the ISS.**

***Keywords*— NASA, microgravity, ferro electric liquid crystals, freely suspended LC films, hydrodynamics**

I. Introduction

Observation and Analysis of Smectic Islands in Space (OASIS) proposes series of experimental study of the interfacial and hydrodynamic behavior of freely suspended liquid crystal (FSLC) films in the microgravity environment on board the International Space Station. FSLC films are formed from rod-shaped molecules that self -organize as bulk materials into fluid smectic LC phases: periodic stackings of layers in which each layer is a two dimensional (2D) fluid on the order of a molecular length in thickness. FSLC films exhibit a combination of physical characteristics that have made them uniquely exciting systems for the study of equilibrium and out-of-equilibrium phenomena in reduced dimensionality, for example liquid crystal ordering and fluctuations in two dimensions, and the effects of finite size on liquid crystal phase transitions.

FSLC films quantized in thickness in integral numbers of layers are the thinnest known stable condensed phase structures and have the largest surface-to-volume ratio of any condensed phase preparation, making them ideal for studies of fluctuation and interface phenomena. The interactions which are operative in liquid crystals are generally weak, leading to the easy manipulation of order by external agents such as applied fields and surfaces, and to significant fluctuation phenomena with extended spatial correlations. These fluctuation, field, and surface effects, combined with the wide variety of LC order parameters and symmetries, makes FSLC films a rich system for probing basic fluid physics.

Based upon extensive ground-based experiments on planar- and spherical bubble FSLC films, we were able to illustrate the possibilities for microgravity study and to develop microgravity experiments. Spherical bubble FSLC films tethered on fine needles can be made with very small ratios of film area to perimeter, yielding systems in which coupling to bulk fluid is extremely weak. These ground-based experiments motivate the proposal of a variety of microgravity studies using the tethered bubble geometry. The proposed experiments will be carried out on bubbles formed and studied in situ in dedicated sample chambers designed in the ISS glovebox system.

During the course of this extensive ground based experiments, we were also able to test some of the vital parts of the proposed OASIS flight hardware on board the parabolic flight aircrafts. These successful preliminary test flights have given us several key information to design and develop much complicated space flight hardware with high confidence.

The major experimental objective is to test in detail of theories of hydrodynamic flow, relaxation of hydrodynamic perturbations, and of hydrodynamic interactions in 2D structure. Microgravity experiments will be pursued with the following objectives:

(1) Freely suspended bubbles in microgravity, without convection, and sedimentation represent nearly ideal, physically and chemically homogeneous 2D fluid systems for the precision study of 2D hydrodynamics. The effects of introducing islands will be studied, both as controllable inclusions that modify the flow and as markers of flow.

(2) Studying the behavior of collective systems of 1D layer step interfaces on 2D bubble surfaces, including the equilibrium spatial organization and interaction of islands, and the nonequilibrium coarsening dynamics of island emulsions.

(3) To carry out the first thermocapillary experiments on homogeneous two dimensional fluids.

(4) To study the dependence of surface tension and line tension on film thickness and Burgers vector.

(5) To probe the effects of a spontaneously broken symmetry in the 2D film surface on the interaction of islands, exploring the stability of topologically stabilized emulsions of 1D interfaces in 2D.

We anticipate that the OASIS flight experiment to be conducted on board the ISS in the early part of 2014. Currently OASIS passed the Requirement Design Review (RDR) phase and we have initiated the design and development of the space flight hardware and will enter the Preliminary Design Review in the early Spring of 2012.

II. SCIENCE BACKGROUND

A. *Background and Research Context*

Smectic liquid crystals are phases formed by rod-shaped molecules organized into one-dimensionally (1D) periodic arrays of layers, each layer being on the order of a molecular length thick. In the fluid smectic phases, the smectics A and C (SmA and SmC), for example, each layer is a two dimensional (2D) liquid, with only short-ranged positional pair correlations within the layer planes and from layer to layer in the direction parallel to the layer planes. **Figure 2.1** shows schematically the structure of these phases and several representative liquid crystal (LC) molecules.

The interactions which are operative in liquid crystals are generally weak in comparison to those in crystalline phases, leading to the facile manipulation of the order and structure of liquid crystals by external agents such as applied fields, surfaces, and flow.

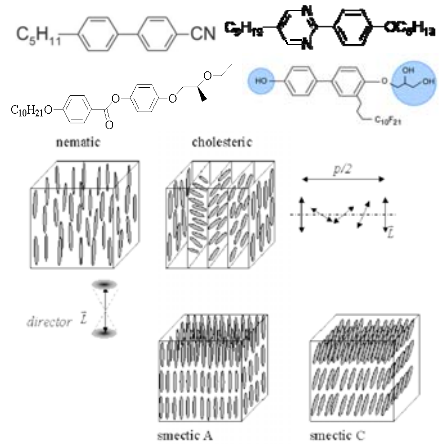


Figure 1.1: Examples of some molecules that form liquid crystals, along with sketches of the four most common liquid crystal phases.

B. *Freely Suspended Liquid Crystal Films*

The modern study of freely suspended thermotropic smectic films was begun in 1973, with the discovery by Noel Clark and coworkers that films of fluid smectics as thin as two molecular layers (thickness, t ~ 6 nm) could readily be made by coating the edge of a hole in a thin plate and drawing a film across the hole with a second plate. The process and resulting structure is sketched in **Figure 2**. The inherent smectic fluid layering on the one hand enables such films to be drawn, and on the other hand stabilizes their structure, enabling films as thin as a single molecular layer be made and studied [**i**]. The smectic layering allows only films that are an integral number N layers thick, quantization that ensures that a film of a particular number of layers will be physically homogeneous with respect to its layer structure over its entire area. Film stability is further enhanced by the absence of any solvent and by component molecular weights typically in the range 200 < MW < 400 Daltons, which make the vapor pressure over the films quite low.

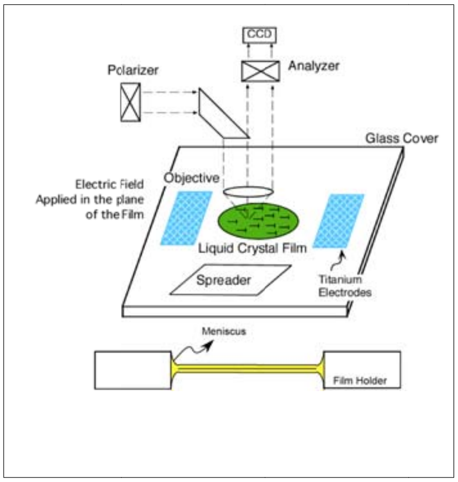


Figure 2: Laboratory setup for making planar freely suspended smectic liquid crystal films. The spreader is used to draw the film over a hole in a flat thin plate. Observation in reflected light visualizes variations in film thickness and the molecular orientation field across the film, with Depolarized Reflected Light Microscopy (DRLM) Electrodes enable the film response to in-plane electric field to be studied. The cross section shows the homogeneous film, stabilized in thickness by the smectic layering connecting to bulk LC in the meniscus.

III PROPOSED MICROGRAVITY EXPERIMENTS

A. Background and Related Research

Here, we present: (**i**) the results of the ground-based research on planar- and spherical bubble (SB) -FSLC films that motivated the development of the OASIS proposal, and (**ii**) the description of the proposed microgravity experiments.

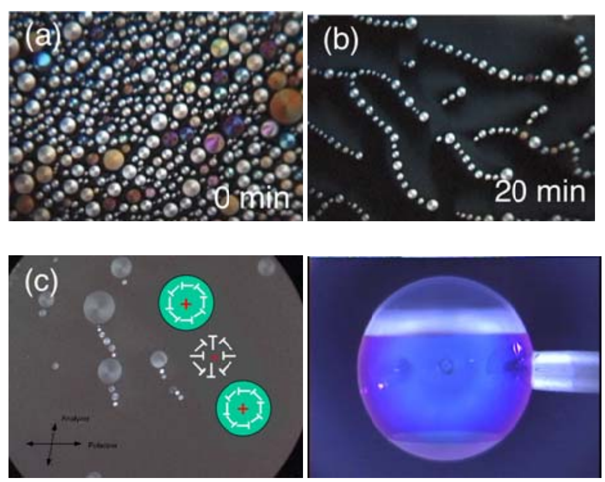
3.1 Key Properties of Freely Suspended Liquid Crystal Films

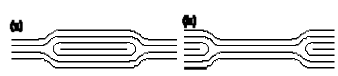
The key relevant FSLC film properties are as follows: (**i**) The smectic layering forces FSLCs to be quantized in thickness in integral numbers of layers (the layer number N) and suppresses pore formation and bursting. This enables the formation in air of stable, single-component, layered, fluid, FSLC films as thin as a single molecular layer (~3 nm thick). (**ii**) The low vapor pressure of smectic-forming compounds enables the long-term stabilization of these films, such that a given few-layer thick film can be studied in the laboratory for many months. Such films are structures of fundamental interest in condensed matter physics. They are the thinnest known stable condensed phase structures and have the largest surface-to-volume ratio of any condensed phase preparation, making them ideal for studies of fluctuation and interface phenomena. (**iii**) The LC layering makes films of uniform layer number highly homogeneous in basic physical properties such as thickness, surface tension, and viscosity, and, away from the edges of the film, completely free of local pinning or other external spatial inhomogeneities. (**iv**) The interactions which are operative in liquid crystals are generally weak, leading to the easy manipulation of order by external agents such as applied fields and surfaces, and to significant fluctuation phenomena with extended spatial correlations. These fluctuation, field, and surface effects, combined with the wide variety of LC order parameters and symmetries, makes FSLC films a rich system for probing basic fluid physics.

3.2Islands on FSLC Films

An important class of OASIS experiments focus on the behavior of islands on FSLC films. Islands are pancake-like stacks of extra layers on an otherwise uniform thickness film, as shown in **Figures 3.**
Islands are of interest as statistical mechanical and dynamical objects because of their very small mass (10^{-12} grams for a 10 μm diameter island), and coupling to the two-dimensional fluid film. The islands respond in a polar way to applied electric field, indicating that they are either charged or significantly more polarizable that the surrounding film. In a microgravity environment the Perrin length, k_BT/mg for an island will be large compared to the bubble size, meaning that in this case the islands will be thermally dispersed over the bubble surface. Interactions between islands can be controlled by applying electric fields or by charging the bubble.

Figure 3: (a) Array of pancake-shaped smectic islands





generated on a ~1 cm diameter smectic C bubble. (b) Island chain aggregates during coarsening process. (c) Islands have topological defect strength +1 in the azimuthal tilt plane orientation field, and each one is accompanied by a -1 defect in the background film. The -1 defects mediate an attractive interaction between the islands. The images are 700 µm wide. (d) An 8 mm diameter smectic bubble tethered on a needle and viewed in reflected light. The needle edge is razor-sharp, making the Plateau border extremely thin. This and the small ratio of needle radius to bubble radius severely restricts transport of material onto or from the bubble (weak tethering limit). The horizontal bands indicate the 1D interfaces (layer steps) separating regions with different layer number N, with the thicker regions sedimenting in the 1g laboratory environment. In microgravity the disposition of such interfaces will be determined by their mutual interactions. (e) Schematic film profiles for islands and pores. We propose to explore the physics mediating the change in layer number.

IV. Motivation for Microgravity Research

DECOUPLING FROM BULK AND THE COLLECTIVE BEAHOIOR OF ONE-DIMENSIONAL INTERFACES

FSLC films exhibit a combination of physical characteristics that have made them uniquely exciting systems for the study of equilibrium and out-of-equilibrium phenomena in reduced dimensionality, for example liquid crystal ordering and fluctuations in two dimensions, and the effects of finite size on liquid crystal phase transitions. Here we show that FSLC films in microgravity present extraordinary opportunities for the study of fluid dynamic and thermodynamic behavior in reduced dimensionality, and for the exploration of fundamental nonequilibrium fluid interfacial phenomena.

FSLC films can be made in planar (**Figures 3.1a,b,c,e**) or spherical bubble (**Figures 3.b**) geometries. With horizontal flat films, near zero-g conditions for in-plane motion can be achieved. However, the behavior of such films, especially with respect film thickness and island dynamics, is controlled to a significant extent by contact with a bulk reservoir of material at the boundary of the film holder, to which it is coupled through a meniscus. The undesirable effects of coupling to the bulk can be effectively eliminated by reducing the perimeter length and volume of the meniscus. Under these conditions, the dynamics of dislocation loops will be dominated by Ostwald ripening and dislocation loop coalescence, and will be constrained by the constant bubble diameter (imposed via a constant Laplace pressure difference between the interior and exterior of the film).

Unfortunately, it is not possible to effectively use this isolated smectic bubble geometry in Earth's gravity. In a terrestrial environment the Perrin length for an island, k_BT/mg, will be small compared to the bubble size, meaning that the islands should sediment to the bottom of the bubble, which is what is observed. In microgravity, on the other hand, k_BT/mg will be large compared to the bubble size and the islands should be thermally dispersed over the bubble surface. *Microgravity conditions provide the only viable route to the creation of large, uniform, boundary-free fluid films for precision studies of defect dynamics and hydrodynamics in two dimensions*.

V. JUSTIFICATION OF EXTENDED DURATION OF MICROGRAVITY ENVIRONMENT

A. Limitations of Terrestrial Experiment

As discussed above, there are a variety of 2D fluid phenomena that are difficult or impossible to study in terrestrial experiments under Earth's gravity. In the typical freely suspended smectic film geometry, the boundary of the film plays a dominant role in dislocation dynamics, providing the driving force for dislocation motion (the Laplace pressure difference between the film and the meniscus) and controlling the rate of dislocation motion (via dissipation due to flow of material through the meniscus). It is also quite difficult to eliminate gravity-driven convection due to thermal gradients in terrestrial experiments, making precision studies of 2D hydrodynamics and thermocapillary effects in smectic films problematic at best.

B. Limitations of Drop Tower and Parabolic Flight Experiments

The amount of time needed for generating smectic bubbles and doing experiments greatly exceeds that available in a drop tower or on aparabolic flight . Ground studies of the generation of bubbles, the creation of island emusions and island coarsening processes indicate a characteristic time scale of about one hour for the islands to substantially merge.

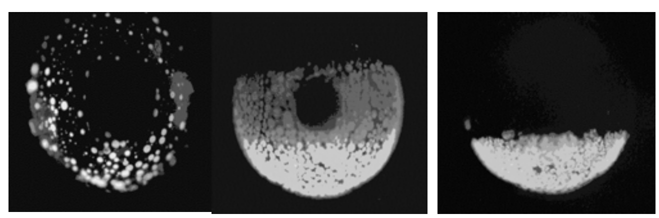


Figure 4 : Islands sedimenting on a freely suspended 8CB smectic bubble. Photographs show smectic C bubbles, ~20 mm in diameter, viewed in reflected light. The dark parts of the bubbles are ~ 3 to 5 smectic layers thick. Pancake shaped islands having additional layers are evident. The black spot in the center of the bubbles is an illumination artifact. In the bubble geometry, the ratio of the Plateau border length around the needle to the film area is very small relative to planar films. This enables the formation of island arrays by a slight decrease of internal pressure. Islands can be dispersed on the film by an air jet (left), but in 1 g sediment to the bottom within a few minutes (right).

VI. SCIENCE OBJECTIVES IN MICROGRAVITY ENVIRONMENT

A. Study of 2D Hydrodynamics

We will pursue detailed tests of theories of hydrodynamic flow, relaxation of hydrodynamic perturbations, and of hydrodynamic interactions in 2D. Freely suspended bubbles in microgravity, without islands, convection, and sedimentation represent nearly ideal, physically and chemically homogeneous 2D fluid systems for the precision study of 2D hydrodynamics. The effects of introducing islands will be studied, both as controllable inclusions that modify the flow and as markers of flow.

B. Probing the Collective Behavior of 1D Interfaces in a 2D Space

To goal will be to study the behavior of collective systems of 1D layer step interfaces on 2D bubble surfaces, including the equilibrium spatial organization and interaction of islands, and the nonequilibrium coarsening dynamics of island emulsions; . In addition to yielding information about a number of relatively weak physical effects (thickness-dependent surface tension and line tension, disjoining pressure, etc.), we anticipate that this will clarify the effects of dimensionality in coarsening dynamics (e.g., on dynamic scaling behavior).

C. Investigation of Thermocapillary Effects

We will carry out the first thermocapillary experiments on homogeneous two dimensional fluids. The proposed microgravity experiments present a unique opportunity to explore thermocapillarity, their translational symmetry and absence of convection, mitigating anomalous effects to the maximum extent possible, enabling detailed studies of the thermocapillarity of 2D fluids.

D. Study of Surface Tension and Line Tension

We will study the dependence of surface tension and line tension on film thickness and Burgers vector. Ground-based experiments indicate that equilibrium and nonequilibrium island behavior should be sensitive to this dependence, enabling critical tests of extant theoretical predictions.

E. Textural Interactions

We will probe the effects of a spontaneously broken symmetry in the 2D film surface (the appearance of 2D polar, XY-like ordering and accompanying electrostatic polarization) on the interaction of islands, exploring the stability of topologically stabilized emulsions of 1D interfaces in 2D.

VII. CONCEPTUAL DESIGN OF THE PROPOSED FLIGHT EXPERIMENT

A. Design Considerations

The experimental apparatus required to observe smectic islands in space will be developed in the MSG of ISS with the design based on proven, ground-based experimental hardware. A conceptual design showing the additional capabilities necessary to accommodate our experiment, is conceptualized in Figure 7.1.

Sample chambers will be equipped for the following functionalities and experiments: (i) Automated bubble inflation to a selected size. (ii) Bubble positioning. (iii) Gas jets tangent to, but not contacting the bubble surface, and tether meniscus heating to generate and study in-plane flow and pull material from the meniscus, generating islands (circular domains with extra layers) on the film. (iv) High-resolution and low resolution imaging of the bubble in reflected light to observe bubble flow and dynamics and coarsening of island emulsions. (v) Electrodes and local heaters near but not contacting the bubble for local application of electric field to the film surface, and for the study of thermocapillary effects. (vi) Optical measurement of film thickness. (vii) Droplet deposition of single islands onto the bubble for the study of weak island interactions. (vii) Dynamic bubble inflation and deflation system and (viii) Temperature control to study the effects of LC phase behavior.

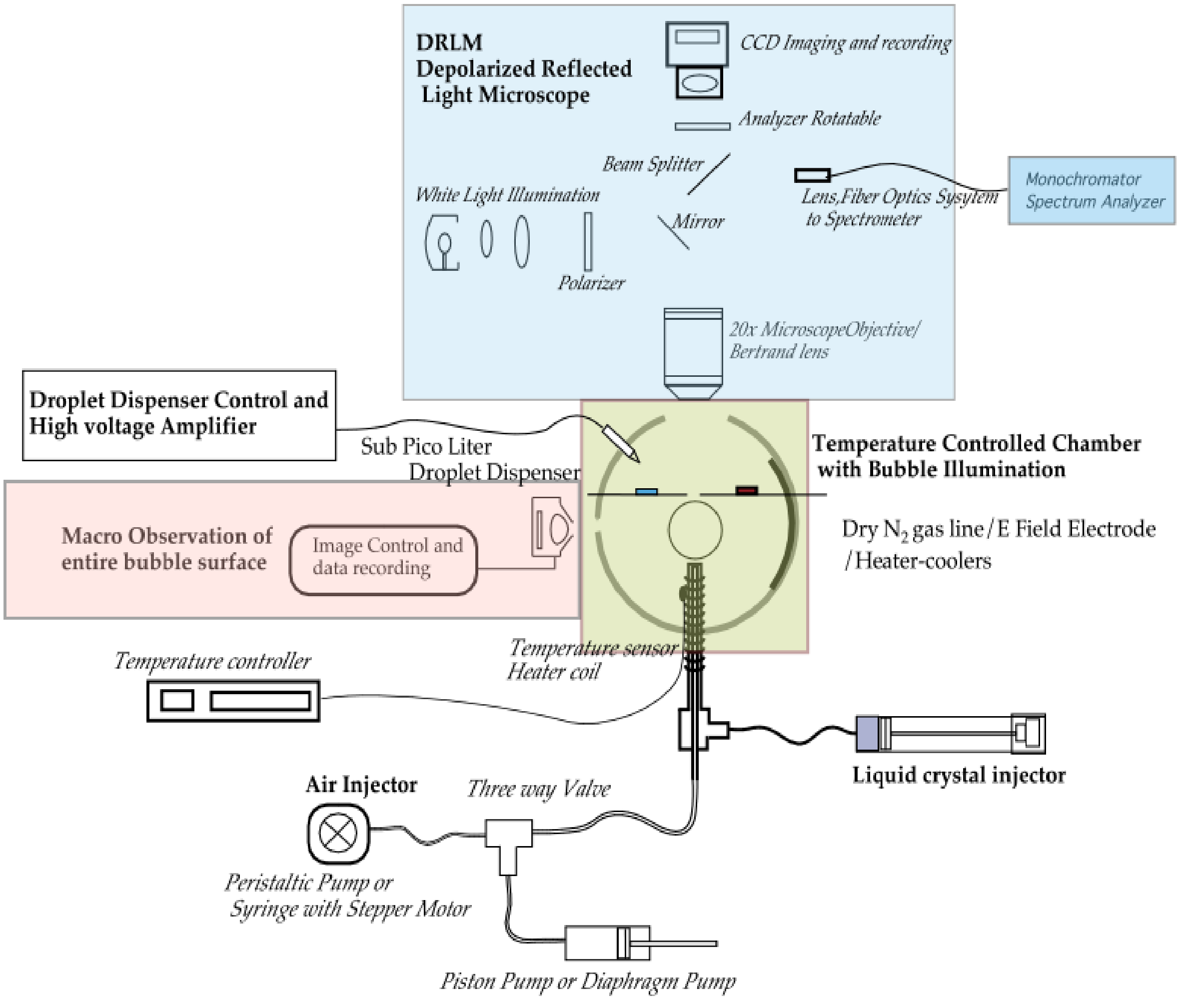


Fig. 5: OASIS experimental concept. Each bubble box contains four independent bubble chambers, which can be individually positioned under the DRLM Depolarized Reflected Light Microscope and the Spectrometer for simultaneous observation using high resolution video microscopy from above and low resolution video from the side. Each chamber includes a CCD camera, and hardware to inflate a bubble, generate islands, measure bubble and island thicknesses, and apply electric fields and temperature gradients

.

Experimental observations of island interactions and dynamics will require both high- and low-resolution video microscopy. Principal processes of interest include coarsening, coalescence, and diffusion of islands, island/hole generation, thermocapillary effects, and island interactions when charged or placed in an external electric field (applied through the shearing needles). These experiments will be performed varying the external stimuli such as magnitude of applied field and induced temperature (gradient), pressure oscillation frequency and quench depth, as well as bubble/island thickness and bubble size.

VIII. CONCLUSION

A. Anticipated Knowledge To Be Gained From Oasis: Value And Applications

[1] 2D Hydrodynamics

The two dimensional hydrodynamics of truly homogeneous two dimensional fluids has never been studied. Freely suspended bubbles without islands represent nearly ideal, physically and chemically homogeneous 2D fluid systems for the precision study of 2D hydrodynamics. In microgravity, the absence of convection and sedimentation enables the study of the hydrodynamics of inclusions (islands) in a nearly ideal 2D fluid. This will enable detailed tests of theories of diffusion, relaxation of hydrodynamic perturbations, and of hydrodynamic interactions in 2D.

[2] Coarsening and Ostwald Ripening Dynamics

3D coarsening dynamics in emulsions, foams, and other nonequilibrium systems is an important and relatively well-studied problem. 2D coarsening dynamics of 1D interfaces is of increasing importance, but detailed studies are complicated by the lack of appropriate model systems. The proposed bubble experiments offer a well-characterized, homogeneous platform for the study of 2D coarsening dynamics. In addition to yielding information about a number of relatively weak physical effects (thickness-dependent surface tension and line tension, disjoining pressure, etc.), we anticipate that this will clarify the effects of dimensionality in coarsening dynamics (e.g., on dynamic scaling behavior).

[3] Thermocapillary Effects

Thermocapillary effects in fluids are notoriously difficult to characterize in Earth's gravity due to the complicating effects of convection and chemical partitioning of components in thermal gradients. Our proposed microgravity experiments with one-component smectic materials will mitigate these effects to the maximum extent possible, and will enable detailed studies of the thermocapillarity of fluid interfaces. These will be the first thermocapillary experiments on homogeneous two dimensional fluids.

[4] Surface Tension and Line Tension

Our ground-based experiments show a subtle dependence of surface tension on film thickness, which is impossible to detect, much less measure, with the standard techniques. This indicates that a possible outcome of the coarsening experiments will be information about the thickness dependence of surface tension. These experiments may also yield information about the Burgers vector dependence of the dislocation line tension, enabling critical tests of extant theoretical predictions.

[5] Perturbing Smectic Bubbles

The suite of proposed perturbation experiments (local heating, pressure quenches, electric field application, non-uniform flow) are relatively simple experiments which, in the context of free-standing smectic bubbles, probe unique physical states and interfacial conditions, enabling the study of hitherto unexplored fluid phenomena.

B. Current Status. Parabolic Flight Experiment Results.

During the course of this extensive ground based experiments, we were also able to test some of the vital parts of the proposed OASIS flight hardware on board the parabolic flight aircrafts. These successful preliminary test flights have given us several key information in regards to the design and develop much complicated space flight hardware with high confidence. During the two parabolic flight campaigns in the year 2010 March and November supported by the German Space agency (DLR) we have designed and tested major components of OASIS parts including the bubble formation, island generation, bubble chamber illumination, and droplet dispensing devices and systems all controlled via the pc. We also tested the pico liter droplet dispensing system using very high speed camera to record the trajectory and droplet size evaluation, as it is necessary that the droplet volume be less than sub pico liter in order to artificially generate islands less than 100 microns to start with. We were able to form droplets of water/glycerol mixture and as well as 8CB liquid crystal. We have obtained several data from these two parabolic flights and data analysis is underway.

After the highly successful completion of the Science Concept Review in September 2008, we have been successfully on going with ground based research at the University of Colorado, Department of Physics, successful vital hardware component tests at the NASA Glenn Research Center and as well as at the Department of Physics, University of Magdeburg. OASIS is currently in the phase of Requirement Design Review expected to be conducted in March 2011.

IX. ACKNOWLEDGEMENT

The authors would like to acknowledge funding supported by the NASA under grant number NNX07AE48G, entitled "Structures and Dynamics of Freely Suspended Liquid Crystal". Support and funding of DLR for the international collaborative research effort for Professor Ralf Stannarius and provision of the parabolic flights is also highly appreciated.

REFERENCES

1. *Handbook of Liquid Crystals*, D. Demus, J. Goodby, G.W. Gray, H.-W. Spiess, V. Vill eds. (Wiley, Weinheim, 1998).
2. "LIGHT-SCATTERING STUDY OF TWO-DIMENSIONAL MOLECULAR-ORIENTATION FLUCTUATIONS IN A FREELY SUSPENDED FERROELECTRIC LIQUID-CRYSTAL FILM," C.Y. Young, R. Pindak, N.A. Clark, and R.B. Meyer, *Physical Review Letters* 40, 773 – 776 (1978).
3. *Defects and Geometry in Condensed Matter Physics* D.R. Nelson (Cambridge, Cambridge, 2002).
4. "DIRECT MEASUREMENT OF ORIENTATION CORRELATIONS: OBSERVATION OF THE LANDAU-PEIERLS DIVERGENCE IN A FREELY SUSPENDED TILTED SMECTIC FILM," D.H. Van Winkle and N.A. Clark, *Physical Review Letters* 53, 1157 – 1160 (1984).
5. "FERROELECTRIC-ANTIFERROELECTRIC PHASE TRANSITION IN A TWO-MOLECULAR-LAYER FREE-STANDING LIQUID-CRYSTAL FILM,"Ch. Bahr and D. Fliegner, *Physical Review Letters* 70, 1842 – 1845 (1993).
6. "SIMULTANEOUS OBSERVATION OF ELECTRIC FIELD COUPLING TO LONGITUDINAL AND TRANSVERSE FERROELECTRICITY IN A CHIRAL LIQUID CRYSTAL," D. R. Link, J. E. Maclennan, and N. A. Clark, *Physical Review Letters* 77, 2237 – 2240 (1996).

7. “FLUCTUATIONS AND CLINICITY IN TILTED SMECTIC LIQUID CRYSTALS,” M.A. Glaser and N.A. Clark, ***Physical Review E*** 66, 021711 (2002).

8. “MANIPULATION OF ISLANDS ON FREELY SUSPENDED SMECTIC FILMS AND BUBBLES USING OPTICAL TWEEZERS,” A. Pattanaporkratana, C. S. Park, J. E. Maclennan, and N. A. Clark,***Ferroelectrics*** 310 131 – 136 (2004), www.e-lc.org.

9. “TOPOLOGICAL INCLUSIONS IN 2D SMECTIC FILMS,“ D. Pettey, T.C. Lubensky, and D.R. Link, ***Liquid Crystals*** 25, 579 – 587 (1998).

10. “SURFACE-TENSION-GRADIENT-INDUCED FLOW IN FREELY SUSPENDED LIQUID CRYSTALLINE FILMS,“ M.I. Godfrey and D.H. Van Winkle, ***Physical Review E*** 54, 3752 – 3764 (1996).

11. “TURBULENCE IN FLOWING SOAP FILMS: VELOCITY, VORTICITY, AND THICKNESS FIELDS, “ M. Rivera, P. Vorobieff, and R.E. Ecke, ***Physical Review Letters*** 81, 1417–1420 (1998).