THRESHOLD PHENOMENA IN SOFT MATTER

by

ZHIBIN HUANG

Submitted in partial fulfillment of the requirements
For the degree of Doctor of Philosophy

Dissertation Adviser: Dr. Charles Rosenblatt

Department of Physics
CASE WESTERN RESERVE UNIVERSITY

May 2008
We hereby approve the thesis/dissertation of

ZHIBIN HUANG

candidate for the Ph.D. degree*.

Charles Rosenblatt
(chair of committee)

Rolfe G. Petshek

Jie Shan

J. Iwan D. Alexander

2/19/2008

*We also certify that written approval has been obtained for any proprietary material contained therein.
I grant to Case Western Reserve University the right to use this work, irrespective of any copyright, for the University’s own purposes without cost to the University or to its students, agents and employees. I further agree that the University may reproduce and provide single copies of the work, in any format other than in or from microforms, to the public for the cost of reproduction.

ZHIBIN HUANG

(sign)
Table of Contents

Table of Contents ......................................................... iv
List of Tables ............................................................. vii
List of Figures ............................................................. viii
Acknowledgement ........................................................... xii
Abstract ............................................................................ xvi

1 Introduction ........................................................................ 1
  1.1 Liquid Crystals ............................................................. 1
    1.1.1 Mesomorphic Phases .............................................. 1
    1.1.2 The Building Blocks of Liquid Crystals ..................... 2
    1.1.3 Classification of the Mesophases .............................. 4
    1.1.4 Order Parameter in Nematics ................................. 10
    1.1.5 Surface-induced Smectic Ordering in Nematics .......... 14
    1.1.6 Free Energy of Nematics ....................................... 15
  1.2 Rayleigh-Taylor Instability .......................................... 19
    1.2.1 What Is the Rayleigh-Taylor Instability ..................... 19
    1.2.2 Theoretical Approaches to the Rayleigh-Taylor Instability ... 22
    1.2.3 Numerical Simulation Methods ............................... 28
    1.2.4 Experimental Methods for the Rayleigh-Taylor Instability ... 37
  1.3 Outline of Experiments .................................................. 39

2 Large Polar Pretilt for the Liquid Crystal Homologous Series Alkyl-
    cyanobiphenyl (nCB) ...................................................... 47
  2.1 Introduction .............................................................. 47
  2.2 Sample Preparation ..................................................... 50
  2.3 Experimental Setup ..................................................... 52
  2.4 Optical Retardation Measurement ................................. 53
  2.5 Dual Easy Axis Model ................................................. 53
  2.6 Pretilt Angle .............................................................. 55
  2.7 Result and Discussion .................................................. 58
3 Smectic Tilt Susceptibility: Anharmonic Behavior in Surface-induced Smectic Layers above the Nematic–Smectic-A Transition Temperature

3.1 Introduction
3.2 Experimental Preparation
  3.2.1 Sample Preparation and Material
  3.2.2 Experimental Setup
3.3 Results
3.4 Theoretical Model
3.5 Data Analysis
3.6 Discussion
3.7 Summary
References

4 Rayleigh-Taylor Instability for Immiscible Fluids of Arbitrary Viscosities: Magnetic Levitation Investigation and Theoretical Model

4.1 Introduction
4.2 Carlès’ Theoretical Model
4.3 Hele-Shaw Cell Construction and Two Working Fluids
  4.3.1 Density Measurement
  4.3.2 Viscosity Measurement
  4.3.3 Interfacial Tension Measurement
  4.3.4 Material Parameters for the Aqueous Mixtures
4.4 Magnetic Levitation
4.5 Data Acquisition and Analysis
4.6 Results and Discussions
4.7 Summary and Prospect
References

5 Rayleigh-Taylor Instability Experiments with Precise and Arbitrary Control of the Initial Interface Shape

5.1 Introduction
5.2 Working Fluids and Experimental Setup
5.3 Data Acquisition and Results
5.4 Initial Perturbation Control and Data Analysis
5.5 Control of the Wire Shape by Calculating the Magnetic Force Profile
5.6 Summary
References
Appendices

A Digitization of the Interface between Two Fluids in an Image 134
  A.1 Algorithm in Canny Edge Detection ........................ 134
    A.1.1 Results ........................................ 138
    A.1.2 Summary ........................................ 139
  References ............................................. 139

B The Measurement of the Relative Magnetic Permeability of a Material 141
  B.1 The Solution to the Boundary Value Problem .............. 141
  B.2 Magnetic Force on a Wire ................................ 144
  B.3 Measurement of Relative Permeability .................... 144
  References ............................................. 147

C Mapping of Images Distorted by Optical Parallax 148
  C.1 Horizontal Correction to the Original Image with Optical Parallax .. 149
  C.2 Vertical Correction ..................................... 152
  C.3 The Image after Optical Parallax Correction .............. 153

D Procedure on Performing the Rayleigh-Taylor Instability Experiment 155
  D.1 Cell Cleaning .......................................... 155
  D.2 Alignment of Two Pole Pieces ................................ 156
  D.3 Setup .................................................. 156
  D.4 Filling with the Fluids ................................... 156
  D.5 Video Acquisition ...................................... 157

Bibliography 158
List of Tables

4.1 Empirical constants for the shape dependent parameter $H$ [127,128]. 97
4.2 Material parameters. ............................................................. 98

5.1 Interface perturbation amplitude $h_{0}$, terminal velocity $\left(\frac{dh}{dt}\right)_{t}$, and growth coefficient $\alpha_{s}$ for different wire amplitudes $A_{w}$. ............. 121
5.2 Growth coefficients for bubbles ($\alpha_{b}$) and spikes ($\alpha_{s}$). ............ 128

C.1 the difference between the real dimension and the vertical length in the image due to the distortion. ................................. 153
List of Figures

1.1 The sketch of an isotropic liquid and a crystalline solid. ............ 2
1.2 Molecule structure of a liquid crystal. .............................. 3
1.3 A Schiff’s base composed of a carbon-nitrogen double bond. ....... 3
1.4 The smectic phase of a lyotropic liquid crystal. ...................... 5
1.5 The sketch of a uniaxial nematic and a biaxial nematic phase. .. 6
1.6 Schematic diagram showing the cholesteric structure. ............... 7
1.7 (a) Twist deformation in a nematic liquid crystal; (b) splay deform-
    ation; (c) bend deformation. ....................................... 8
1.8 The layered structure of smectic-A and smectic-C. .................. 9
1.9 Schematic of the interface between a nematic liquid crystal and a
    substrate. ................................................................. 15
1.10 An example of the Rayleigh-Taylor instability. ..................... 20
1.11 Two incompressible fluids having different densities. The unper-
    turbed interface is the plane $z = 0$. ............................ 23
1.12 A small disk in a plane tangent to a surface passing through point $A$. 30
1.13 A fluid system moving in a velocity field. .......................... 33

2.1 Two basic alignments: homeotropic alignment and planar alignment. 48
2.2 Chemical structure of SE-1211. ....................................... 50
2.3 A schematic of rubbing a slide on a wedge ............................. 51
2.4 A sample with gradient rubbing on a substrate. ..................... 51
2.5 A schematic of the experimental setup. ............................... 52
2.6 Retardation as a function of rubbing strength at different tempera-
    tures ($\ast: T = 25^\circ; \Box: T = 32^\circ$). .......................... 54
2.7 $\theta_0$ vs $n_f$: Circles correspond to 5CB (open circles at 25 °C and solid circles at 32 °C), stars to 6CB, squares to 7CB, and triangles to 8CB [upward triangles at 33.75 °C (about 0.25 °C above $T_{NA}$), rightward triangles at 34°C, leftward triangles at 36°C, and downward triangles at 38°C]. Typical horizontal error bars are shown, and are due primarily to the uncertainty in locating $n_f = 0$. Uncertainty in $\theta_0 \sim \pm 2^\circ$, primarily from the spread in $\theta_0$ due to small variations in rubbing strength.

3.1 The change of $\theta_s$ on cooling. ........................................ 66
3.2 A schematic of cell with liquid crystal mixture SCE12R. ............. 70
3.3 A sketch of the experimental setup. ...................................... 70
3.4 Intensity vs voltage $V_{cell}$ applied to the cell at selected temperatures of ■ 87, □ 86, • 85, ○ 84, ▲ 83, △ 82, ▼ 81. ......................... 72
3.5 Intensity vs $V_{cell}$. Scale enlarged by a factor of (a) 200 and (b) 8000 relative to Fig. 3.4. Solid lines show examples of calculated intensity, and arrows show the threshold voltage at several temperatures. Note that $V_{LC}$ was calculated from the applied voltage $V_{cell}$. Symbols correspond to ○ 84, ▲ 83, △ 82, and ▼ 81 °C. ......................... 73
3.6 $V_{cell}^{th}$ vs reduced temperature $T - T_{NA}$. Note that the threshold vanishes above $T = T_a$, corresponding to approximately 85.5 °C. .......... 78
3.7 Dielectric constants $\varepsilon_\perp$ and $\varepsilon_{\parallel}$ vs reduced temperature $T - T_{NA}$. .... 78
3.8 Bend elastic constant $K_{33}$ vs reduced temperature $T - T_{NA}$. .... 79
3.9 $W_{2eff}$ (solid circles, left-hand scale) and $W_{4eff}$ (open circles, right-hand scale) vs reduced temperature $T - T_{NA}$. Solid line is a fit of $W_{2eff}$ to the form $W_2 + A[(T - T_{NA})/T_{NA}]^{-3\nu}$ (see text). .... 80
4.1 Schematic of Hele-Shaw cell. ........................................... 88
4.2 Pipette calibration with the distilled water. Apparent volume is the reading on the pipette. .................................................. 94
4.3 Pipette calibration with chloroform. .................................... 94
4.4 Schematic of a setup for viscosity measurement. The picture of the U-shaped viscometer is taken from the manufacturer [126]. .......... 94
4.5 A photo of the experimental setup for the interfacial tension measurement. A pipette is used to produce a droplet of a heavy fluid in a light fluid. A green laser is used to induce fluorescence of dye in the light fluid for illumination of the interface between two fluids. .. 96
4.6 A pendant droplet for the interfacial tension measurement. ......... 97
4.7 A schematic of magnetic levitation. .................................... 100
4.8 Magnetic field $H_x$ (right axis) and $H_x \partial H_x / \partial z$ (left axis) as functions of position $z$ when the separation between two pole pieces is 1.7 cm. Note that $z = 0$ corresponds to the position of closest approach of the pole pieces and the quantity $H_x \partial H_x / \partial z$ is maximum at $z = -0.8$ cm.

4.9 The characteristic decay time of the magnetic field. Note that the response time of the magnetic force is a half of that of the magnetic field (=118 ms).

4.10 A schematic of experimental setup for the measurement of the characteristic decay time.

4.11 Images of interfacial region (not the entire cell) at time (a) 0.67 s, (b) 2.00 s, (c) 2.33 s, and (d) 2.67 s after field is turned off. Aqueous solution is 56.4 wt.-% MnCl$_2$ · 4H$_2$O in water. The interfaces fitted from the digitized images are shown by the dashed lines. Horizontal and vertical scales are the same. Bright spot is a reflection from the light source.

4.12 Four-parameter function applied to fit the interface for all times.

4.13 Two-parameter function applied to refit the interface for all times.

4.14 Dimensional growth rate $\bar{\sigma}$ for fastest growing perturbation vs Ca. Open triangles represent experiment and solid triangles theory.

4.15 Dimensionless growth rate $\sigma$($=\frac{d(\log A)}{dt}t_{ref}$), (open circles) and dimensionless wave vector $k$ (open squares) for fastest growing perturbation, vs Ca. The two theoretical curves for each parameter correspond to the extreme values of $\mu^2$ [0 (solid line) and 1 (dotted line)]. Error bars for $k$ and $\bar{\sigma}$ due to fittings are approximately the size of the data symbols ($\pm$5%).

5.1 Refractive index and density of the aqueous mixtures. Note that the refractive index $n$ was measured by using a Nd:YaG laser beam at 532 nm.

5.2 Cartoon of experimental setup. Note that no wires are attached to the outside of the cell, and that the x-axis view is expanded for clarity. The arrow indicating the direction of $\nabla (H^2)$ applies only in the absence of the wires.

5.3 Cartoon of experimental setup. Note that wires are attached to the outside of the cell, and that the x-axis view is expanded for clarity. The arrow indicating the direction of $\nabla (H^2)$ applies only in the absence of the wires.

5.4 A measure of the initial growth rate $\sigma^*$. 

x
5.5 Images of spike growth vs. time in sec., where \( t = 0 \) is the time at which the magnet current first begins to decrease. Column a) no wire; b-d) wires of amplitude \( A_w = 0.1, 0.2, \) and 0.3 cm, respectively.

5.6 Main figure: \( \frac{\vec{F}_{\text{wire}} - \vec{F}_0}{\vec{F}_0} \) in the midplane of the cell for \( A_w = 0.25 \) cm. Wide gray curve is projection of wire into the midplane, dotted line is fluid interface at \( t = 0 \), and solid curve is actual fluid interface measured at \( t = 0.265 \) s. The force, which affects fluid 1 due to its large \( \chi_1 \), is largest near the wire’s crossing points, and results in a collection of fluid 1 at the crossing points. Inset: Dimensionless interface perturbation amplitude \( k h_{k0} \) vs. wire amplitude \( A_w \).

5.7 Two earliest resolvable interface amplitudes fitted with Eq. (5.3) to obtain \( h_{k0} \) for wire amplitude \( A_w = 0.25 \) cm.

5.8 Growth data vs. time for cell with wire of amplitude \( A_w = 0.3 \) cm. a) the position of the front, b) the velocity of the front, and c) the instantaneous growth coefficient \( \alpha_s \).

5.9 Growth data vs. time for cell without wires. a) the position of the front, b) the velocity of the front, and c) the instantaneous growth coefficient \( \alpha_s \).

5.10 Comparison of measurement techniques for the growth rate coefficient \( \alpha \). Courtesy to Cabot \textit{et al.} [129] for this figure where \( \tau \) is a natural timescale.

A.1 A frame from a video.

A.2 An edge profile obtained after performing an edge detection.

A.3 The original image with an edge profile.

B.1 an infinite cylinder in a uniform magnetic field.

B.2 a schematic of the experimental setup for relative permeability measurement.

C.1 the original image with optical parallax.

C.2 the original image with labeled vertical lines and a symmetric axis in bold line.

C.3 a trapezium in the raw frame mapped onto a rectangle in the processed frame.

C.4 the image without the horizontal distortion.

C.5 the new image without optical parallax.

D.1 A schematic of the cell between magnets.
First of all, I would like to thank my advisor Prof. Charles Rosenblatt for his insightful instruction, and for his support for motivating me to do all the research work, and for his answers to my questions. His guidance and encouragement have been invaluable to me. I feel very lucky and very proud to have him as my advisor.

I am very grateful to Prof. Pierre Carlès at Université Paris 6/CNRS. I can’t forget his theoretical model on the RT instability experiments in Hele-Shaw regime and his precise prediction on the interfacial tension between Chloroform and MnCl₂·4H₂O aqueous mixture. It is his theoretic prediction that leads us to the right direction and eventually to a success in our experiments. Although the distance between Paris and Cleveland is pretty far, (thanks to the internet) our communications are very fast and frequent.

I would like to thank Prof. Rolfe Petschek, Prof. J. Iwan D. Alexander, and Prof. Jie Shan for being on my thesis committee.

I would also like to thank many professors such as Walter Lambrecht, Harsh Mathur, Philip Taylor, Kenneth Singer, John Ruhl and Jie Shan. I learned a lot from their classes. I really appreciate their work.

During my graduate study at Case, I had a lot of fun when discussing homework problems with my classmates Guilin, Minhua, Pavel, Aditi, Heifei, Lei, Greg, and Soloman. I would like to thank them for sharing their experience on solving physics problems.

I would also like to thank Mary MacGowan, Patricia Bacevice, Lori Morton, Sue
Rischar, and Betty Gaffney for helping me do some paperwork.

I am grateful to my labmates. When I joined our research group, Neha was writing her thesis. Although she was very busy, she showed me how to prepare \( \text{MnCl}_2 \cdot 4\text{H}_2\text{O} \) aqueous mixture and to perform liquid bridge experiments using magnetic levitation technique. Minhua gave me a training on sample preparation including breaking a big piece of glass into small slides, cleaning glass slides, coating polyimide on the slides, and baking the slides. She also showed me how to measure a cell thickness using interferometric scheme technique. Ishti helped me a lot in using Igor program to do data acquisition and in creating a controllable pattern on polyimide surface using AFM. He also showed me how to prepare PMMA solution. Gianni joined our group after two months I worked in our group. He brought to our group some fresh experimental methods along with a lot of fun. One thing I won’t forget is that he introduced MATLAB to me. Although I used to make a program using C/C++ or FORTRAN, I decided to switch to MATLAB because it is very powerful in the signal processing and the graphics. Later on, I felt comfortable with the video analysis in our fluid dynamic experiments. On the other hand, I should mention that I had a good time when I played “Ping Pong” together with Gianni and Minhua (Note that “Ping Pong” is an informal name of the table tennis. One interesting thing is its explanation one guy gave me when I played “Ping Pong” in the Veal fitness center. He said that the term “Ping Pong” originated by aristocrats is used for those amateurs to have fun and the name of table tennis is for the professional players to earn a reputation). Normally, we take it granted that Italian
are good at soccer but not at “Ping Pong”. However, it is unbelievable that Gianni is very good at “Ping Pong”.

One year later, Ruiting joined our group. Chinese became a majority in our group. Sometimes we made a fun of each other in Chinese. When we joked about democracy, you may imagine what’s so fun. For example, if it would be applied to our group, then the minority such as Chuck, Ishti, and Gianni would have to speak Chinese since our group had four Chinese at that moment (I forgot to mention that Lei joined our group in the Fall semester of 2004).

In Summer 2006, Valentin spent several months working as a visiting Ph.D candidate in our group. He is easygoing. It was fun to work together with him on the phase separation using silicone oil mixed with liquid crystal E$_7$. In the early 2007, he came back to our group working as a postdoctoral fellow (Congratulations on his success in his graduate study!). In the mean time, Antonio and Tim joined our group. I would like to thank them for working together with me about the RT instability experiments. Also, I would like to thank Daeseung for his suggestions on my job hunting and his useful discussions about the silicone oil droplet experiments and the RT instability experiments.

Without soccer, I believe that my life would totally be different. But it is soccer that makes me become social. I had a lot of fun when I played soccer games with colleagues at Case. I have to thank many people for their organization of and contribution to some wonderful games, which made my graduate study rich and varied.
I would also like to express many thanks to many brothers and sisters in Cedar Hillcrest Baptist Church where I began to realize the endless love from God.

Finally, I would like to thank my mother who has always supported my studies. Also, I couldn’t find any words to express my gratitude to my wife for her instant support to me and for her time and her patience in taking care of my son during my graduate study when I spent most of my time in my research projects at Case. When I feel frustrated, she always stays with me. I am very proud of having a lovely son Andrew who gives me a lot of fun when we play together. Without their support, I would not have been able to make anything as far as I have.
Threshold Phenomena in Soft Matter

Abstract

by

ZHIBIN HUANG

Although two different fields are covered, this thesis is mainly focused on some threshold behaviors in both liquid crystal field and fluid dynamic systems.

A method of rubbed polyimide is used to obtain pretilt. Sufficiently strong rubbing of a polyimide (SE-1211) results in a large polar pretilt of liquid crystal director with respect to the homeotropic orientation. There exists a threshold rubbing strength required to induce nonzero pretilt. For the homologous liquid crystal series alkyl-cyanobyphenyl, we found that the threshold rubbing strength is a monotonic function of the number of methylene units. A dual easy axis model is then used to explain the results.

Fréedericksz transition measurements have been used to determine the quadratic and quartic coefficients associated with the molecules’ tilt with respect to the layer normal in surface-induced smectic layers in the nematic phase above the smectic-A–nematic phase transition temperature. Both the quadratic and quartic coefficients are consistent with the scaling relationship as predicted in theory,
and their ratio is approximately constant.

A Rayleigh-Taylor instability experiment is performed by using a magnetic field gradient to draw down a low density but highly paramagnetic fluid below a more dense fluid in a Hele-Shaw cell. When turning off the magnetic field, the RT instability occurs \textit{in situ} and the growth of the most unstable wavevector is measured as a function of time. The wavelength of the RT instability along with the growth rate was measured as a function of capillary number (which is related to the density difference and interfacial tension between two fluids). A theory for the instability that permits different viscosities for two immiscible fluids was developed, and good agreement was found with the experimental results. The technique of magnetic levitation promises to broaden significantly the accessible parameter space of gravitational interfacial instability experiments.

A method is introduced to control initial conditions for a Rayleigh-Taylor instability, wherein a dense fluid sits metastably atop a less dense fluid, a configuration that can be stabilized using a magnetic field gradient when one fluid is highly paramagnetic. On switching off the magnetic field, the instability occurs as the dense fluid falls under gravity. By affixing appropriately shaped magnetically-permeable wires to the outside of the cell, arbitrarily-chosen and well-controlled initial perturbations are imposed on the interface. Using this technique, the perturbation amplitudes, growth rates, nonlinear growth coefficients, and terminal velocities are obtained to examine both the linear and nonlinear growth regimes.
Chapter 1

Introduction

1.1 Liquid Crystals

1.1.1 Mesomorphic Phases

We all remember learning that matter exists in three common states: solid, liquid and gas. However, this is not the whole story about the states of a matter. The term liquid crystal or mesophase refers to phases with intermediate phases between the isotropic liquid and crystalline solid states. The familiar phase of liquid and solid are shown in Fig. 1.1. As can be seen, in the isotropic liquid the molecules are completely random or disordered and free to move around with only short range order, whereas in the crystalline solid the molecules’ long axes tend to be parallel and confined to a regular lattice structure.

Between 1850 and 1888 researchers found that some materials behaved strangely [1]. For instance, in 1850, W. Heintz reported that stearin melts from a solid to a cloudy liquid at 52 °C, changes to an opaque liquid at 58 °C and transitions to a clear
liquid at 62.5 °C. A state of liquid crystal, an intermediate between the crystalline solid and amorphous liquid, was not discovered until in 1888 when an Austrian botanist named Friedrich Reinitzer experimented with a cholesterol substance and noted that it had two melting points [2].

The properties of liquid crystals have been exploited in many devices, especially in liquid crystal displays.

1.1.2 The Building Blocks of Liquid Crystals

Liquid crystalline phases are formed by using anisotropic objects, either elongated or disk-like. These anisotropic materials includes organic molecules viewed as short rigid rods, or long helical rods, polymers and more complex units with associated structures of molecules and ions.

In general, the molecules of commonly occurring liquid crystals are aromatic and have the basic structures shown in Fig. 1.2. They are comprised of a side chain R,
two or more aromatic rings A and A’ connected by linkage groups X or Y, and a terminal group R’ at the other end. The side chains and terminal groups can be alkyl\((C_nH_{2n+1})\), alkoxy \((C_nH_{2n+1}O)\), acyloxy, alkylcarbonate, alkoxy carbonyl, and the nitro and cyano groups, among others [3, 4].

Examples of the aromatic rings are saturated cyclohexane or unsaturated phenyl, biphenyl, and terphenyl. If the liquid crystal molecules contain benzene rings, they are referred to as benzene derivatives, which are the majority of liquid crystals. The others include heterocyclics, organometallics, sterols, and some organic salts or fatty acids.

The physical and optical properties of liquid crystals are related to the properties of the constituent groups. For instance, the central linkage group determines the chemical stability of liquid crystals. For example, Schiff-base liquid crystals are unstable and Fig. 1.3 shows a Schiff’s base. These constituent groups and their various combinations govern elastic constants, viscosities, transition temperatures, anisotropies and optical nonlinearities.
1.1.3 Classification of the Mesophases

The mesophases may be classified into two categories [5]: disordered crystal mesophases and ordered fluid mesophases. The constituent molecules of disordered crystal mesophases are arranged upon a lattice with orientational freedom, and form plastic crystals. The constituent molecules of ordered fluid mesophases undergo long-ranged orientational order and also may have some long-ranged translational order. These are referred to as liquid crystals. The orientation ordering of the liquid crystals arises because the constituent molecules are very rigid and anisotropic in shape, also anisotropic van der Waals interacts. It is an essential requirement for the formation of liquid crystals or mesomorphism to occur that the molecule must be highly geometrically anisotropic in shape like a disc or a rod. While long thin (rod-like) molecules forming calamitic liquid crystals, thin flat (disk-like) molecules form discotic liquid crystals, which have been discovered relatively recently [6].

Liquid crystal phases may occur in a pure compound as the temperature is changed. These are termed thermotropic liquid crystals. When liquid crystal phases occur in the presence of a solvent such as water, they are referred to as lyotropic liquid crystals. Fig. 1.4 shows a smectic phase of a lyotropic liquid crystal formed by dissolving sunset yellow food dye in water.

According to the nomenclature proposed by Friedel [7], thermal liquid crystals are classified into three types: nematic, cholesteric and smectic. This thesis shall treat two types: nematic and smectic.
The simplest liquid crystalline phase is the nematic. The word “nematic” invented by G. Friedel is from the Greek “nema” meaning thread because the threadlike appearance of domains can be observed under the polarizing microscope. Molecules in a nematic phase spontaneously tend to orient with their long axes roughly parallel and on average point in a preferred direction called director $\mathbf{n}$, as shown in Fig. 1.1.

There are several types of nematic phases, but all of them can be thought of as liquids with long-ranged orientational ordering although the average orientation may change spatially as the nematic phase can sustain an elastic distortion. For most nematic liquid crystals a rotation around the director does not make any difference and these liquid crystals are consequently referred to as uniaxial as shown in Fig. 1.5(a); some liquid crystals exhibit asymmetry with respect to rotations around the preferred axis of alignment and are referred to as biaxial, with the building blocks
Figure 1.5: The sketch of a uniaxial nematic and a biaxial nematic phase.

shaped in the form of rectangles as shown in Fig. 1.5(b). A biaxial phase was first observed in lyotropic systems [8].

An important type of nematic liquid crystals is made up of molecules that are chiral or absent of inversion symmetry. As human hands lack mirror symmetry, this property is often referred to as handedness. A cholesteric is composed of unequal numbers of left- and right-handed molecules, with a helical structure of a pitch defined as the distance it takes for the director to rotate one full turn in the helix [9], as seen in Fig. 1.6. These liquid crystals have a variety of applications. For instance, since these materials are able to selectively reflect light of wavelengths equal to the pitch length, a specific color would be reflected when the pitch equals the corresponding wavelength of light in the visible spectrum. Based on the temperature dependence of the gradual change in director orientation between successive layers, the pitch is modified, resulting in an alteration of the wavelength of reflected light according to the temperature. Thus, by using mixtures of different types of these
Figure 1.6: Schematic diagram showing the cholesteric structure.

Liquid crystals sensors are created with a wide variety of responses to temperature change.

A nematic liquid crystal will undergo a deformation when there exists an external perturbation field. An example is shown in the Fig. 1.7(a), which describes a "solid" subjected to a torsional stress by which the molecules are translationally displaced. Such a deformation called a twist deformation simply involves the rotation of molecules in the direction of the torque. Other types of deformations such as splay and bend are shown in Fig. 1.7(b) and (c), respectively. These two deformations involve mainly changes in the director $n$. 
The term smectic was invented by G. Friedel is from the Greek “σμη γαμα” meaning soap. Smectic phases are found at lower temperatures than nematics. Unlike nematics, smectic liquid crystals have positional order in one direction, that is to say, molecules’ position is correlated in some ordered pattern. From a structural point of view, smectic phases have layered structures with an interlayer spacing that can be exploited by X-ray diffraction. Smectic liquid crystals are more ordered than nematics, exhibiting both long-range positional and orientational ordering in the molecular arrange. In most smectic liquid crystals, molecules are mobile in two directions and can rotate around their axis while the smectic layers can easily slide over one another, such that a partial fluid property is possessed by smectic liquid crystals, although with higher viscosity than nematic liquid crystals.

There are many different types of smectic phases. The simplest smectic phase is smectic-A (Sm-A). In smectic-A phases, the molecules are oriented along the layer normal as shown in Fig. 1.8(a). The essential feature of smectic-A phase can be summarized as follows [10]:

- It has a layer structure with the orientation of molecules orthogonal to the
There is no long-range ordering of the center of gravity inside each layer and each layer is a two-dimensional liquid.

- It is optically uniaxial, with the optical axis pointing along the layer normal.

- Like the nematic where $n$ and $-n$ are equivalent, the directions $z$ and $-z$ are equivalent.

To obtain a constant layer spacing, it is required to impose the condition $n \cdot \nabla \times n = 0$ for all macroscopic deformations of smectic A. Thus it is easy to see that the twist and bend deformation allowed in nematics is forbidden within smectic-A.

There exist strong thermal fluctuations of layers in Sm-A phase. As a consequence the positional ordering is not really long-range (due to the Landau-Peierls instability). In other words, the mean-square displacement of the layers diverges in the thermodynamic limit and the long-range order is destroyed [10–12]. In practice, since the logarithmic growth of the fluctuations with the sample size is very slow
and difficult to be directly observed in the typical experimental sample [13], the positional ordering can be treated as truly long-range [11,14].

A smectic-C (Sm-C) phase has lower symmetry than smectic-A and so tends to occur at lower temperatures. In smectic-C phases, the molecules are tilted on average at some angle with respect to the layer normal, as shown in Fig. 1.8(b). Sm-C is optically biaxial.

### 1.1.4 Order Parameter in Nematics

In a transition between phases, some physical component of the system changes in symmetry and the physical quantity that changes may be described by some suitable parameters. Order parameters are fundamental to the description of phase transitions. Since the free energy is a function of the temperature and the order parameter, first-order phase transitions have a discontinuity in the first derivative of the free energy with a thermodynamic variable such as temperature. Second-order phase transitions have a continuity in the first derivative of the free energy but a discontinuity in the second derivative of the free energy [15,16].

The nematic phase has a lower symmetry than the isotropic liquid and generally occurs at lower temperature. In other words, the nematic phase is more ordered than the high-temperature isotropic liquid. An order parameter is defined to explain this ("more ordered") quantitatively. The order parameter can be very different for different cases: in the case of ferromagnetic transitions the order parameter is the magnetization which is a vector with three independent components [10]; in the case
of liquid-vapor transitions, the order parameter is the density difference between the liquid and vapor phases and is a scalar.

### 1.1.4.1 Microscopic Order Parameter

Considering rigid rods which are the simplest type of objects allowing nematic behavior, the nematic axis $\mathbf{n}$ is taken as the $z$-axis of the $(x, y, z)$ laboratory frame. The axis of one rod is labeled by a unit vector $\mathbf{a}$ with its polar angle $\theta$ and azimuthal angle $\phi$.

\[
\begin{align*}
a_x &= \sin \theta \cos \phi \\
a_y &= \sin \theta \sin \phi \\
a_z &= \cos \theta
\end{align*}
\]

A distribution function $f(\theta, \phi)d\Omega$ giving the probability of finding the rods in a small solid angle ($d\Omega = \sin \theta d\theta d\phi$) can be used to describe the state of alignment of the rods. Note that in conventional nematics

- $f(\theta, \phi)$ is independent of $\phi$ since the phase has complete cylindrical symmetry around $\mathbf{n}$.
- $f(\theta) = f(\pi - \theta)$ since the directions $\mathbf{n}$ and $-\mathbf{n}$ are equivalent.

As a result of the above properties, in order to characterize the alignment not through $f(\theta)$, but by a related numerical parameter, a term with higher multipoles is required. The first multipole giving non-trivial answer is the quadrupole and then
the order parameter is defined as

\[ S = \frac{1}{2} \langle (3\cos^2\theta - 1) \rangle = \int f(\theta)\frac{1}{2}(3\cos^2\theta - 1)d\Omega. \]

(1.1)

If \( f(\theta) \) is strongly peaked at \( \theta = 0 \) and \( \theta = \pi \), then \( S = 1 \) for parallel alignment. On the other hand, if \( f(\theta) \) were peaked around \( \theta = \pi/2 \) (perpendicular alignment), \( S \) would be \(-1/2\). For the random orientation where \( f(\theta) \) is independent of \( \theta \), \( \langle \cos^2\theta \rangle = \frac{1}{3} \) and \( S = 0 \). For liquid crystals the typical value of the order parameter ranges from 0.3 to 0.9 and is temperature-dependent [5,10]. Thus \( S \) is a measure of the alignment.

For molecules without rotational symmetry, a more general tensor order parameter \( S_{ij} \) is required and defined as [4,10]

\[ S_{ij} = \frac{1}{2} \langle 3(n \cdot i)(n \cdot j) - 1 \rangle, \]

(1.2)

where \( i, j, k \) are unit vectors along the molecular axes. The three diagonal components are given by

\[ S_{ii} = \frac{1}{2}(3\sin^2\theta\cos^2\phi - 1) \]
\[ S_{jj} = \frac{1}{2}(3\sin^2\theta\sin^2\phi - 1) \]
\[ S_{kk} = \frac{1}{2}(3\cos^2\theta - 1) \]

Note that \( S \) is traceless tensor since \( S_{ii} + S_{jj} + S_{kk} = 0 \).
1.1.4.2 Macroscopic Order Parameter

The order parameters defined previously in terms of the alignment of states can be translated into expressions in terms of the anisotropic parameters such as magnetic, electric and optical susceptibilities. For instance, in terms of the magnetic anisotropy $\Delta \chi = \chi_{||} - \chi_{\perp}$, a so-called macroscopic order parameter to characterize the bulk response can be defined as $[4,10]$

$$Q_{\alpha\beta} = G \left( \chi_{\alpha\beta} - \frac{1}{3} \delta_{\alpha\beta} \sum_{\gamma} \chi_{\gamma\gamma} \right), \quad (1.3)$$

where $\alpha, \beta = x, y, z$ and $\delta_{\alpha\beta}$ the Kronecker delta. $G$ is the normalization constant. To be more explicit, consider a uniaxial material, choosing the $z$-axis parallel to the nematic axis such that the tensor $\chi_{\alpha\beta}$ has the form as

$$\chi_{\alpha\beta} = \begin{pmatrix} \chi_{\perp} & 0 & 0 \\ 0 & \chi_{\perp} & 0 \\ 0 & 0 & \chi_{||} \end{pmatrix}. \quad (1.4)$$

Writing $Q_{\alpha\beta}$ explicitly in terms of the diagonal components, we have

$$Q_{\alpha\beta} = \begin{pmatrix} \frac{2}{3} \left( \chi_{\perp} - \chi_{||} \right) & 0 & 0 \\ 0 & \frac{1}{3} \left( \chi_{\perp} - \chi_{||} \right) & 0 \\ 0 & 0 & \frac{2}{3} \left( \chi_{||} - \chi_{\perp} \right) \end{pmatrix}. \quad (1.4)$$
1.1.4.3 Relationship between Microscopic Order Parameter and Macroscopic Order Parameter

The relation between the macroscopic tensor $\chi_{\alpha\beta}$ and the microscopic quantities $S_{ij}$ defined in Eq. (1.2) can be found as follows [10]

$$\chi_{\alpha\beta} = c \sum_{ij} A_{ij} \langle (\mathbf{n} \cdot \mathbf{i}) (\mathbf{n} \cdot \mathbf{j}) \rangle = c \sum_{ij} A_{ij} \langle i_{\alpha} j_{\beta} \rangle,$$

where $A_{ij}$ is the magnetic polarizability tensor of one molecule and can be derived through the study of the diamagnetism in the crystalline phase. For uniaxial rods, the relation was simplified by Saupe and Maier as

$$\chi_{||} - \chi_{\perp} = c (A_{||} - A_{\perp}) S(T). \quad (1.5)$$

Here order parameter $S(T)$ is temperature-dependent. The difference $(A_{||} - A_{\perp})$ depending on the detailed structure and rigidity of molecules can be derived from magnetic resonance measurements [17].

1.1.5 Surface-induced Smectic Ordering in Nematics

When the nematic liquid crystal is placed in contact with a substrate, a surface (or surface of substrate) is created to limit the nematic phase [18]. The presence of such a limiting surface introduces a perturbation of nematic order over a distance $\xi$ near the surface, as seen in the Fig. 1.9(a). $\xi$ is a few molecular length and defines the thickness of the transition region between the substrate and the nematic liquid phase for the system far away from any phase transition of the bulk. The thickness
Figure 1.9: Schematic of the interface between a nematic liquid crystal and a substrate.

can increase and reach a macroscopic value when the liquid crystal approaches to a bulk phase transition.

The perturbation induced by the substrate arises from the symmetry breaking induced by the substrate. That is to say, the translation invariance of the nematic phase is broken by the presence of the surface, resulting in a tendency of a layer of molecules at the surface. In other words, the center of mass of the molecules near to the surface has the tendency to be located in a plane parallel to the surface. The existence of this layer would cause the formation of a second one and so on, thus inducing a positional ordering of the molecules at the interface between the nematic phase and the substrate.

As the molecules are perpendicular or tilted at an angle with respect to the layer normal, the ordering at the interface induces smectic order locally.

1.1.6 Free Energy of Nematics

The following free energy terms are frequently referred in this thesis.
1.1.6.1 Landau-de Gennes Free Energy

The Landau-de Gennes approach is applied to weakly first-order nematic-isotropic phase transitions. The free energy expansion for uniaxial molecules is given by [3, 5, 19]

\[ f_s = \frac{1}{2}a(T - T^*)S^2 - \frac{1}{3}bS^3 + \frac{1}{4}cS^4 + O(S^5), \]

where \( T^* \) is the virtual second-order transition temperature and terms higher than the forth-order are ignored. For a nematic liquid crystal of rod-like molecules we have \( b > 0 \) and the first order transition temperature \( T_c = T^* + \frac{2b^2}{9ac} \).

1.1.6.2 Elastic Distortion Free Energy

Because a twist deformation simply involves the rotation of molecules in the direction of the torque and the center of gravity of molecules is not displaced, the elastic energy involved in the twist deformation is quite small. Other types of deformations such as splay and bend shown in Fig. 1.7(b) and (c), respectively, involve mainly changes in the director \( n \), thus the elastic energy changes in the latter two deformations are generally bigger than that in twist deformation.

Following the curvature elasticity theory first developed by Frank [20], the distortion free-energy density associated with these three types of deformations is given by

\[ f_d = \frac{1}{2}K_{11} \left( \nabla \cdot n \right)^2 + \frac{1}{2}K_{22} \left( n \cdot \nabla \times n \right)^2 + \frac{1}{2}K_{33} \left( n \times \nabla \times n \right)^2, \]

where \( K_{11}, K_{22}, K_{33} \) are referred as Frank elastic constants and are associated with
three types of deformations: splay, twist and bend, respectively.

Considering a frequently used assumption, namely, the one-constant approximation where $K_{11} = K_{22} = K_{33} = K$, the distortion free-energy density becomes

$$f_d = \frac{1}{2}K \left[(\nabla \cdot n)^2 + (\nabla \times n)^2\right].$$  \hspace{1cm} (1.8)

1.1.6.3 Anchoring Energy

Surface effects have an influence on the bulk nematic phase with the order recovered at a distance from the surface greater than $\xi$ shown in Fig. 1.9(a). Close to the interface the molecules on average orient along the fixed orientation labeled with $a$ which is called the anchoring “easy axis” of the nematic liquid crystal at the interface. This orientation is imposed on the bulk molecules due to the elastic forces which tend to orient all the molecules parallel to each other. This phenomenon is so-called anchoring of the nematic liquid crystal at the interface. The angle between the fixed direction $a$ and the surface normal $z$ is called “pretilt” at the interface in Fig. 1.9(a). Note that here “pretilt” is equivalent to the polar angle $\theta$ at the interface in Fig. 1.9(b). For $\theta \simeq 0^\circ$ the alignment of liquid crystals is called “homeotropic”. For $\theta \simeq 90^\circ$ the alignment is called “planar”.

When the orientation of the molecules of nematic liquid crystals at an interface departs from the anchoring direction, there would be some energy cost called anchoring energy.
The polar anchoring energy is expressed as follow [21]

\[ f_\theta = \frac{1}{2} W_\theta \sin^2 \theta, \]  
\[(1.9)\]

where \( W_\theta \) is the polar anchoring strength coefficient. \( W_\theta \) is positive for homeotropic alignment. Considering the high order coefficients, the general anchoring energy is

\[ f_\theta = \frac{1}{2} W_2 \sin^2 \theta + \frac{1}{4} W_4 \sin^4 \theta + \cdots \]  
\[(1.10)\]

The azimuthal anchoring energy is given by

\[ f_\phi = \frac{1}{2} W_\phi \sin^2 \phi, \]  
\[(1.11)\]

where \( W_\phi \) is the azimuthal anchoring strength coefficient which is generally a function of the polar angle \( \theta \).
1.2 Rayleigh-Taylor Instability

Among many types of recognized fluid instabilities exists the instability dealing with two fluids of different densities sitting one over another. Two cases are distinguished as follows:

- the interface of two fluids is accelerated by an impulsive force (the Richtmyer-Meshkov (RM) instability);

- fluids are accelerated towards each other with a time-independent force (the Rayleigh-Taylor instability).

For both cases, the acceleration field is directed from the heavier fluid to the lighter one. Here let us focus on the Rayleigh-Taylor instability.

1.2.1 What Is the Rayleigh-Taylor Instability

The Rayleigh-Taylor (RT) instability occurs when a density gradient is subjected to an acceleration by an antiparallel pressure gradient in the presence of perturbations ($\nabla \rho \cdot \nabla p < 0$). An example is when a dense fluid is placed atop a less dense fluid in the presence of gravity, and falls when the interface is perturbed. These perturbations evolve into spikes of heavy fluid and bubbles of light fluid which penetrate into both fluids. The evolution of the RT unstable interface is very complex, involving the phenomena such as the formation and detachment of droplets, competition among rising bubbles, and entrainment and turbulent mixing of a finite layer of fluid.
In order to clearly illustrate this instability, consider a very good example introduced by Sharp [22]. A ceiling of a room is plastered uniformly with water to a depth of one meter, as shown in Fig. 1.10. The water layer will fall. This is not due to the lack of the supporting pressure. In fact, the air pressure equivalent to that of a column of water ten meters thick is sufficient enough to support the water layer against ceiling. However, the atmosphere fails to constrain the water/air interface to flatness. It is due to the unstable interface and gravity that the water layer falls. No matter how carefully the water layer was prepared, it will deviate from the initial plane by a small amount. When a portion of the fluid lies higher than the mean surface, it experiences higher pressure than is needed for its support: it begins to
rise. While a neighboring portion of the fluid is hanging a little lower than the mean surface, it will require more than the average pressure for its support: it starts to fall. In this way small disturbances of the interface grow.

The RT instability can be divided into three stages. The first stage of the instability is dominated by the small amplitude growth of perturbation relative to the wavelength of the perturbation ($kh \ll 1$). In this phase, the amplitude of perturbation exponentially grows as a function of time. According to the linear theory [23], the growth rate depends on the fluids' density ratio, viscosity, surface tension and compressibility. When the perturbation grows to more than 10-40% of its wavelength [22], the linear theory is no longer appropriate. The RT instability transitions to the second stage characterized by nonlinear perturbation growth with bubbles of the light fluid rising into the heavy fluid while the heavy fluid penetrates into the light fluid as spikes. As the instability continues, the bubbles interact with each other while the mushroom-shaped spikes of the heavy fluid occur. Finally, the interaction evolves into chaotic or turbulent mixing which dominates the third stage of the RT instability. In this final stage, there are some interesting phenomena such as bubble competitions and breakup of spikes by various mechanisms.

The importance of the RT instability is in many fields such as astrophysics [24,25]. The RT instability plays an important role in the formation of supernova remnant, during which the expanding heavy gas is decelerated by the surrounding light gas, and the RT instability occurs on the interface between the heavy and light gases, resulting in the mixing of materials.
The RT instability is important for some technological applications as well. An example is inertial confinement fusion (ICF) [24–28]. In ICF, the fuel of deuterium-tritium fusion is compressed by the expanding light gas surrounding the fuel to get high temperature. In this process, the RT instability occurs at the interface between the fuel and the light gas, inducing the mixing of materials, which subsequently causes a severe degradation of the capsule performance by seeding the deceleration-phase RT instability and preventing the onset of the ignition process, and thus reduces the temperature. Hence, for the success of ICF, it is very crucial to fully understand the nature of the RT instability such as the RT growth rate.

1.2.2 Theoretical Approaches to the Rayleigh-Taylor Instability

1.2.2.1 Linear Theory

In 1900, Rayleigh investigated a linear analysis of the initial stage of the Rayleigh-Taylor instability for an ideal incompressible fluids [29]. Late in 1950, Taylor developed the linear theory for the case of uniformly accelerated fluid layers of finite thickness. The linear theory was extended by Richtmyer [30] to his impulsive model for the Richtmyer-Meshkov instability. In this linear theory, the fluids are assumed to be incompressible and to have uniform densities, while the flow is assumed to be two-dimensional and irrotational.

Let us consider two fluids of different densities under gravity, as shown in Fig. 1.11. Since the interface between two fluids is unstable when the denser fluid sits
atop the lighter one, the perturbed interface is given by

\[ z = a(t) \cos(ky), \quad [a(t)/\lambda \ll 1] \quad (1.12) \]

where the amplitude of the perturbation \( a(t) > 0 \) is a function of time, \( k > 0 \) is a wave number, and \( \lambda \) is the perturbation wavelength. Thus the interface is composed of a set of crests and troughs parallel to the \( x \) direction.

According to the theory [23], the amplitude of perturbation is determined by

\[ \frac{d^2a(t)}{dt^2} = \Sigma^2(k)a(t), \quad (1.13) \]

with

\[ \Sigma^2(k) = \left( \frac{\rho_H - \rho_L}{\rho_H + \rho_L} \right) gk - \left( \frac{\gamma}{\rho_H + \rho_L} \right) k^3, \quad (1.14) \]
where \( g \) is the gravitational acceleration, \( \gamma \) the interfacial tension between two fluids, \( \Sigma \) the dimensional growth rate.

The solution to Eq. (1.13) for the fluids with an initially flat interface is

\[
a(t) = a(0) \cosh(\Sigma t).
\]  

(1.15)

Some conclusions can be drawn from Eq. (1.14) as follows:

- \( \Sigma \) is real and the interface is unstable if the interfacial tension \( \gamma = 0 \);

- Interfacial tension stabilizes perturbations shorter than a critical wavelength \( \lambda_c = 2\pi [\gamma/g(\rho_H - \rho_L)]^{1/2} \);

- The relationship between the fastest growing or most unstable wavelength and the critical wavelength is given by \( \lambda_M = \sqrt{3}\lambda_c \).

The linear analysis can be generalized to include other physical effects such as compressibility, density gradients, nonuniform accelerations, shocks and so on. However, the theory is valid only for the initial phase of the instabilities where \( ka \ll 1 \), beyond which the linearized Navier-Stokes equations no longer apply.

1.2.2.2 Nonlinear Regime

After an initial phase of the exponential growth, the disturbances of the interface continue to grow and nonlinear effects dominate the dynamics of the interface: the motion of the interface goes to a nonlinear regime, causing the growth rate to decline [31–34]. In the late nonlinear stage, the interface has a quite complicated form: the
heavy fluid penetrates into the light one, forming long narrow spikes, and the light fluid forms long wide bubbles rising into the heavy fluid.

In nonlinear regime, the shape of interface is very complex and the description of motion of the interface is very difficult. This problem captured Fermi’s attention. In 1951, Fermi [35] proposed a model for the evolution of a bubble and a spike in the late nonlinear stage of the RT instability. He considered the free surface dynamics of one fluid of infinite depth. To describe the motion of the interface, he made an approximation by replacing the rippled surface with a square wave profile which is characterized by three parameters: the heights of the spike and bubble, and the width of the spike. The conservation of mass provides one relation between these parameters. Since this relation reduces the degrees of freedom by one, the dynamics of two degrees of freedom is used to describe the motion of the square wave profile. This dynamics is determined by a variational principle of least action, resulting in information of the asymptotic behaviors of the spike and bubble in the late nonlinear regime. From this model some conclusion can be drawn: the model correctly describes some features of the RT instability with the spike growing as $t^2$ and becoming narrower while the bubble is getting wider, although the correct asymptotic speed of the bubble cannot be given.

Later on Fermi and Neumann [36] extended the model by considering the motion of interface between two fluids with difference densities, and obtained the asymptotic behaviors of the spike and bubble, which are similar to those observed in the case of one fluid. Even if their model doesn’t predict the correct asymptotic speed of the
bubble, it describes the shape of the interface by using a simple function and the dynamics of the function is determined by a variational principle.

Attempts to model the motion of the bubble in the RT instability have been made by several workers. Based on the assumption that the bubble tip has a parabolic shape, Layzer [37] obtained an approximate solution interpolating between the initial small amplitude motion and the late time (steady-state) motion, and gave a successful description of the evolution of a single-mode RT bubble over all the time. Layzer’s work was extended by Hecht et al. [38] to a potential flow model for the motion of the bubbles for the RM instabilities. Their model successfully predicts that in a uniform gravitational field a single bubble attains a constant asymptotic velocity of the bubble, and in a zero gravitational field the velocity of the bubble of wavelength $\lambda$ decays as $\lambda t^{-1}$. However, they failed to describe the entire surface. Later on, Sohn [39] extended the potential flow model to bubbles for both the RM and the RT instabilities, obtaining asymptotic growth rates in good agreement with numerical results. Zhang [40] generalized the potential flow model to spikes for the RM and the RT instabilities, predicting that the asymptotic growth rate of a spike is constant and initial-condition dependent.

There are a number of studies modeling the motion of bubbles rising under gravity by using a steady solution of a free boundary problem. In the frame moving with a bubble, the shape of the bubble varies slowly. This observation suggests that a steady solution may exist in the free boundary problem of fluids. Several authors investigated the motion of bubbles using this approach. For instance, Birkhoff
and Carter [41] investigated “plane bubbles” (2D axi-symmetrical bubbles) rising between parallel walls. They described the problem using nonlinear integral equations and obtained unique solutions to these equations. The similar problem was discussed with the prediction of the existence of one-parameter family of steady solutions characterized by the dimensionless speed [42–44].

While progress in understanding completely the single-bubble problem has been made, research on the more complicated phenomena of bubble interactions is still going on. A phenomenological diffusion approach to theoretical modeling of mixing-layer growth was first introduced by Belenki and Fradkin [45] and developed by several authors [46]. These diffusion models explain well experimental data by using some empirical constants. Statistical bubble model based on bubble dynamics was introduced by Gardner et al. [47] and Glimm et al. [48].

Self-similarity plays a very important role in the evolution of the RT mixing layer involving bubble interactions. The evolution of bubbles complies with the following law: $h(t) = cGt^2$, where $h$ represents the width of the mixing layer, $G$ the acceleration and $c$ an approximate universal constant (about 0.058-0.065 for 2D system and about 0.073-0.077 for 3D system). Many efforts have been made by several authors to derive a self-similarity evolution law. For instance, Chandrasekhar [23] included damping effects such as viscosity and surface tension in his model and obtained a finite wave number of maximum growth. It is because of the existence of the strong spatial damping of short-wavelength modes that longer wavelengths become more competitive during the time evolution, leading to a self-similar evolution law [49].
A single-mode model introduced by Hull [49] simply illustrates mixing motions in the RT instability. In this model, the particle trajectories of a single internal RT mode are described in terms of a stream function (Note that the definition of the stream function can be found in Ref. [50]—“For two-dimensional flows it is possible to eliminate the continuity equation from the system of equations by using only functions which satisfy the continuity equation. This elimination is a formal step toward a solution, and the functions which affect this elimination are the stream functions”). Considering conservation of both the mass and total energy of the system, a linear growth rate expression is obtained, which is consistent with the result predicted by the linear theory in the limit of weak density gradient [49], although the model gives no consistent dynamic description of nonlinear evolution.

1.2.3 Numerical Simulation Methods

To deal with fluid dynamics, it is inevitable to have to resort to computational techniques to study and solve complex problems in fluid flow and heat transfer. The development of computational fluid dynamics (CFD) began with the advent of the digital computers in the early of 1950s. Finite element methods (FEM) and finite difference methods (FDM) are basic tools used to solve partial differential equations in general cases. In FDM, derivatives in the governing equations are written in finite difference forms. Meshes must be structured in either two or three dimensions (for instance, curved meshes have to be transformed to orthogonal cartesian coordinates) so that finite differential equations can be written on structured cartesian meshes.
However, in FEM the unstructured meshes and complex geometries can be easily adopted. By combining FEM with FDM, finite volume methods (FVM) were developed and become popular in recent years because of their simple data structure [51]. FVM are also called control volume methods and are formulated from the product of a unit function with a governing partial differential equations. They have the flexibility for unstructured grids. Another advantage of FVM is that the governing equations retain their physical interpretation rather than distorting the physics due to numerical discrimination of the derivative terms. In addition to FDM, FVM and FEM, available methods such as Monte Carlo methods (MCM), particle-in-cell (PIC) methods, Eulerian and Lagrangian methods, and boundary element methods (BEM) are used in numerical simulation of the governing equations for impressible viscous flows—known as the incompressible Navier-Stokes equations of motion.

1.2.3.1 The Navier-Stokes Equations of Motion

Before the Navier-Stokes equations are addressed, it is necessary to introduce some concepts such as the stress, the stress tensor and the material time derivative.

The Stress

The term stress is referred to as stress at a point. Consider a surface passing through point $A$ in a fluid, while a plane tangent to the surface at the point $A$. A small disk is drawn on this plane and has the outer normal $n$, as shown in Fig. 1.12. The average stress on the surface is defined as follow:

$$T = \frac{F}{S},$$  

(1.16)
where $S$ is the disk area and $F$ is the force acting on its surface $S$. The stress at point $A$ is defined as the limit of $T$ as $S$ tends to zero. Stress is a vector depending on the vector $\mathbf{n}$. The component of the stress normal to $\mathbf{n}$ is called normal stress and that perpendicular to $\mathbf{n}$ is called shear stress.

**The Stress Tensor**

Surface forces acting at a point $A$ on a plane depend on the location of the point and on the plane orientation, as well as on the direction of its outer normal $\mathbf{n}$. It is clear that the forces depend on the coordinates of the considered point. As we know, in cartesian coordinates there are nine stress components $T_{ij}$ (representing a stress in $j$ direction on a face normal to $i$ axis), which can be expressed in a form of $3 \times 3$ matrix as follow [50]:

$$
T = \begin{pmatrix}
T_{11} & T_{12} & T_{13} \\
T_{21} & T_{22} & T_{23} \\
T_{31} & T_{32} & T_{33}
\end{pmatrix},
$$

(1.17)

where the subscripts 1, 2, 3 stand for $x$, $y$, $z$, respectively.

The operator $T_{ij}$ (corresponding to the off-diagonal elements when $i \neq j$) is
called a tensor. It is also called the stress tensor since it is related to a stress vector $T$.

**The Material Time Derivative**

In the study of motion in continuum mechanics the rates of changes in time of quantities vary from one particle to another. These quantities can be density, velocity and acceleration, and may be expressed as functions in a material form. The time rate of their change is meaningful, but with different meanings for different quantities. For instance, we have two different systems: one is a metal ball located at the point $(x, y, z)$ in the lab frame and the other is sphere of fluid located at the same point in another set of coordinates. Let us consider the time rates of change in time of densities of both systems. The notation $d\rho/dt$ can be used to describe the time rate of change of the ball density. For a moving fluid this notation could mean the time rate of change of density at a different point $(x_1, y_1, z_1)$ brought about since a fluid at the time $t + \Delta t$ is different from that at the time $t$. On the other hand, this notation could be the time rate of change of density at the point $(x_1, y_1, z_1)$ at time $t$ but went somewhere at $t + \Delta t$. Obviously, this notation is not clear when it is used in fluid mechanics. Therefore, the total time derivative $D\rho/Dt$ is introduced to describe the time rate of change of density of the fluid system (*system in the thermodynamic sense*), leading to the definition of the material time derivative.

Assuming that a fluid particle at the local position $(x, y, z)$ is moving with a local velocity

$$v = u\hat{i} + v\hat{j} + w\hat{k},$$  

(1.18)
the time rate of density of the fluid system is as follow

\[
\frac{D\rho}{Dt} = \frac{\partial \rho}{\partial t} + \frac{\partial \rho}{\partial x} \frac{\partial x}{\partial t} + \frac{\partial \rho}{\partial y} \frac{\partial y}{\partial t} + \frac{\partial \rho}{\partial z} \frac{\partial z}{\partial t},
\]  
(1.19)

which can be written in vector notation as

\[
\frac{D\rho}{Dt} = \frac{\partial \rho}{\partial t} + u \frac{\partial \rho}{\partial x} + v \frac{\partial \rho}{\partial y} + w \frac{\partial \rho}{\partial z} = \frac{\partial \rho}{\partial t} + \mathbf{v} \cdot \nabla \rho. 
\]  
(1.20)

For time-independent incompressible flows \(\partial \rho/\partial t = 0\), Eq. (1.20) becomes

\[
\frac{D\rho}{Dt} = \mathbf{v} \cdot \nabla \rho, 
\]  
(1.21)

while for the case of a static fluid, it becomes

\[
\frac{D\rho}{Dt} = \frac{\partial \rho}{\partial t}. 
\]  
(1.22)

For a vector quantity \(\mathbf{A}\) the material time derivative can be defined as

\[
\frac{D\mathbf{A}}{Dt} = \frac{\partial \mathbf{A}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{A}. 
\]  
(1.23)

Reynolds Transport Theorem

As shown in Fig. 1.13, a thermodynamic system has the volume \(V_t\) at time \(t\). Consider some general property \(B\) of the system inside \(V_t\), the time rate of change of \(B\) is given by replacing \(\rho\) with \(B\) in Eq. (1.20):

\[
\frac{DB}{Dt} = \frac{\partial B}{\partial t} + (\mathbf{v} \cdot \nabla) B. 
\]  
(1.24)
If we let \( b \) be the value of \( B \) per unit mass in volume \( V_t \), then we have

\[
B = \int_V (\rho b) dV. \tag{1.25}
\]

Eq. (1.24) becomes

\[
\frac{DB}{Dt} = \frac{\partial}{\partial t} \int_V (\rho b) dV + \int_V (\mathbf{v} \cdot \nabla) (\rho b) dV \tag{1.26}
\]

or

\[
\frac{D}{Dt} \int_V (\rho b) dV = \int_V \frac{\partial}{\partial t} (\rho b) dV + \int_S \rho b (\mathbf{v} \cdot \mathbf{n}) dS, \tag{1.27}
\]

where \( \mathbf{n} \) is the unit vector normal to the differential area \( dS \). These two equations (1.26-1.27) state that the rate of change \( B \) for a given system is attributed to the change of \( B \) inside a volume \( V_t \) plus the net outflow of \( B \) through the boundaries of the volume. They are known as the Reynolds transport theorem.

**The Equation of Continuity**

According to the law of conservation of mass, a thermodynamic system has a constant mass \( m \) at all the times. With substitution of \( m \) into Eq. (1.27) (by setting \( b \) to be 1) and \( \nabla \cdot \mathbf{v} = 0 \) (since incompressible irrotational motions are subject to
\( \nabla \cdot \mathbf{v} = 0 \) and \( \nabla \times \mathbf{v} = 0 \) [49]), we have

\[
0 = \frac{Dm}{Dt} = \frac{\partial}{\partial t} \int_V \rho dV + \int_S \rho (\mathbf{v} \cdot \mathbf{n}) dS = \int_V \left[ \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) \right] dV. \tag{1.28}
\]

The above equation has to be satisfied by any system chosen in a domain of the fluid. That is to say, the volume \( V \) can be arbitrary. Thus, we obtain

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0, \tag{1.29}
\]

which is known as the equation of continuity written in terms of the flow field.

**Newton’s Second Law of Motion**

According to Newton’s second law of motion, the rate of change of the momentum of a thermodynamic system is equal to the total forces (body forces and surface forces) acting on the system. Therefore, the change of the momentum of the system can be described as [50,52]

\[
\frac{D}{Dt} \int_V \rho \mathbf{v} dV = \int_V \rho \mathbf{g} dV + \int_S \mathbf{T} dS, \tag{1.30}
\]

where \( \mathbf{g} \) is a general body force per unit mass which has units of acceleration and \( \mathbf{T} \) is the stress on the boundary of the system. One example of \( \mathbf{g} \) is the gravitational acceleration. Consider \( x \)-component of Eq. (1.30), with substitution of the material time derivative, we have

\[
\int_V \left[ \rho \left( \frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} + w \frac{\partial u}{\partial z} - g_x \right) - \left( \frac{\partial T_{xx}}{\partial x} + \frac{\partial T_{yx}}{\partial y} + \frac{\partial T_{zx}}{\partial z} \right) \right] dV = 0, \tag{1.31}
\]
Similarly, the equations for the $y$- and $z$-components are obtained. Because these equations in terms of the three components of the momentum must be satisfied by any thermodynamic system chosen in the domain of the fluid, $V$ is arbitrary and thus the integral must vanish at any point in the domain of the integration. Therefore, we have the following equations:

\[
\rho \left( \frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} + w \frac{\partial u}{\partial z} \right) = \rho g_x + \frac{\partial T_{xx}}{\partial x} + \frac{\partial T_{yx}}{\partial y} + \frac{\partial T_{zx}}{\partial z}, \quad (1.32)
\]

\[
\rho \left( \frac{\partial v}{\partial t} + u \frac{\partial v}{\partial x} + v \frac{\partial v}{\partial y} + w \frac{\partial v}{\partial z} \right) = \rho g_y + \frac{\partial T_{xy}}{\partial x} + \frac{\partial T_{yy}}{\partial y} + \frac{\partial T_{zy}}{\partial z}, \quad (1.33)
\]

\[
\rho \left( \frac{\partial w}{\partial t} + u \frac{\partial w}{\partial x} + v \frac{\partial w}{\partial y} + w \frac{\partial w}{\partial z} \right) = \rho g_z + \frac{\partial T_{xz}}{\partial x} + \frac{\partial T_{yz}}{\partial y} + \frac{\partial T_{zz}}{\partial z}. \quad (1.34)
\]

**Deformation Tensor**

In our daily life, there are a lot of things related to the deformation of materials. For instance, when a fluid is forced to flow along a pipe with a decreasing diameter, it undergoes the action of stress. The deformation caused by the stress is called strain. In general form, strain is a symmetric tensor given by [14]

\[
\epsilon_{ij} = \frac{1}{2} \left( \frac{\partial r_i}{\partial x_j} + \frac{\partial r_j}{\partial x_i} \right), \quad (1.35)
\]

where $r$ represents the displacement field of the fluid system.

The rate of deformation tensor (strain rate) is given by [50]

\[
\dot\epsilon_{ij} = \frac{1}{2} \left( \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right). \quad (1.36)
\]
The Navier-Stokes Equations

The Newtonian fluid is defined as one that satisfies [50]

\[ T_{ij} = -p\delta_{ij} + 2\mu\varepsilon_{ij} = -p\delta_{ij} + \tau_{ij}, \quad (1.37) \]

where \( \delta \) is the Kronecker delta, \( \mu \) the dynamic viscosity and \( p \) the pressure introduced for a moving fluid and defined for an incompressible fluid as \( p = -(T_{xx} + T_{yy} + T_{zz})/3 \), \( \tau_{ij} \) the dynamic viscous stress.

Combining Eqs. (1.32, 1.33, 1.34) and Eqs. (1.36,1.37), we have

\[ \rho \left( \frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} + w \frac{\partial u}{\partial z} \right) = \rho g_x - \frac{\partial p}{\partial x} + \mu \left( \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2} \right), \quad (1.38) \]

\[ \rho \left( \frac{\partial v}{\partial t} + u \frac{\partial v}{\partial x} + v \frac{\partial v}{\partial y} + w \frac{\partial v}{\partial z} \right) = \rho g_y - \frac{\partial p}{\partial y} + \mu \left( \frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2} + \frac{\partial^2 v}{\partial z^2} \right), \quad (1.39) \]

\[ \rho \left( \frac{\partial w}{\partial t} + u \frac{\partial w}{\partial x} + v \frac{\partial w}{\partial y} + w \frac{\partial w}{\partial z} \right) = \rho g_z - \frac{\partial p}{\partial z} + \mu \left( \frac{\partial^2 w}{\partial x^2} + \frac{\partial^2 w}{\partial y^2} + \frac{\partial^2 w}{\partial z^2} \right). \quad (1.40) \]

These three nonlinear second order partial differential equations are called the Navier-Stokes equations. They can be written in their vectorial form as

\[ \rho \frac{Dv}{Dt} = \rho g - \nabla p + \mu \nabla^2 v = \rho g - \nabla p - \mu \nabla \times (\nabla \times v). \quad (1.41) \]

Assuming that two fluids are both viscous, incompressible, and immiscible, the governing equations for unsteady, viscous, incompressible, immiscible two-fluid system and the incompressibility condition are given as follows [53–56]:

\[ \frac{\partial \rho Y_m}{\partial t} + \frac{\partial \rho Y_nv_j}{\partial x_j} = \frac{\partial}{\partial x_j} \left( \rho D_F \frac{\partial Y_m}{\partial x_j} \right) \quad (m = 1, 2), \quad (1.42) \]

\[ \frac{\partial \rho v_i}{\partial t} + \frac{\partial \rho v_i v_j}{\partial x_j} = \rho g_i - \frac{\partial p}{\partial x_i} + \frac{\partial \tau_{ij}}{\partial x_j}, \quad (1.43) \]
where

\[ \tau_{ij} = \mu \left( \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial v_k}{\partial x_k} \right), \]

\( Y_m \) is the mass fraction of species \( m_i \), \( v_{(i,j,k)} = (u, v, w) \) are the components of the velocity \( v \), \( D_F \) is the Fickian diffusivity and \( g_i = (0, 0, -g) \) is the acceleration.

### 1.2.3.2 The Euler Equations

When the viscosity \( \mu \) is set to zero in the vectorial form of the Navier-Stokes equations (1.41), a form of the so-called Euler equations is obtained:

\[ \rho \frac{Dv}{Dt} = \rho g - \nabla p. \quad (1.44) \]

In fact, the Euler equations were formulated earlier than the Navier-Stokes equations. Since the Euler equations cannot generally satisfy the velocity boundary conditions near a rigid boundary, they don’t form a good approximation. However, far from the boundary the Euler equations play an important role in approximations and are generally easier to solve than the full Navier-Stokes equations.

### 1.2.4 Experimental Methods for the Rayleigh-Taylor Instability

There are several different experimental methods to study the RT instability. One method is to accelerate a tank containing a fluid (two fluids) to create an unstable interface between a fluid and air (between two fluids). The first successful experiment to study the RT instability was performed by Lewis [57] using an apparatus for
accelerating a tank containing fluids having different densities vertically downward at accelerations of the order of 50g. Emmons et al. [58] produced an unstable interface between a liquid and air by accelerating a tank on rails. The first important measurements of the growth of a mixing layer were performed by Read [59] using a drop tank accelerated downward by rocket motors. This experiment setup provided a high acceleration and thus a capability of study on the RT instability for a large range of Atwood number. In a similar method, Dimonte and Schneider [60] used the linear electric motor facility to accelerate a drop tank, generating an unstable interface between fluids.

Another method is to turn rapidly upsidedown a tank with a light fluid sitting on the top of a heavy fluid. An example is that Andrews et al. [61] overturned a tank containing two fluids of different densities.

When a solid plate is introduced to separate two fluids in a tank, a configuration with a heavy fluid above a light fluid is initially created. Linden et al. [62] and Dalziel et al. [63] generated an unstable interface by withdrawing the plate from a tank containing a heavier fluid above a lighter fluid. Ramaprabhu and Andrews [64] used a water channel facility to set up an unstable RT flow due to a temperature gradient between two water streams entering through separate inlets. These two streams were separated by a splitter plate with a knife edge at its end. When the cold and hot streams left the edge of the splitter plate, an unstable interface between two streams was produced.

The above methods inevitable have difficulty in obtaining controllable initial
conditions of the interface. As a magnetic levitation technique was introduced in the study of the RT instability, such problems were solved immediately. Pacitto et al. [65] performed the RT instability experiment in a magnetic field, using magnetic fluids (also called ferrofluids) which are colloidal suspensions of magnetic nanoparticles. In their experiment, a new external parameter provided by an applied magnetic field can stabilize or destabilize the fluid interface. Rannacher and Engel [66] recently proposed a theoretical model of the RT instability involving a magnetic field. An initial interface with a magnetic fluid superimposed on a non-magnetic liquid of lower density was achieved by using a spatially homogeneous magnetic field rotating in the plane of the undisturbed interface. In our RT experiments using magnetic levitation technique [67], we deal with classical fluids instead of ferrofluids. On the whole, this method provides very well-controlled initial conditions for the RT instability experiments.

1.3 Outline of Experiments

This thesis is focused mainly on the threshold behavior of liquid crystals and complex fluids, such as Fréederisckz transition; threshold rubbing strength; and perturbations of the interface between two fluids under gravity. The plan of the thesis is as follows.

A method of rubbed polyimide is used to obtain pretilt in chapter 2. Sufficiently strong rubbing of a polyimide (SE-1211) results in a large polar pretilt of liquid crystal director with respect to the homeotropic orientation. There exists a threshold
rubbing strength required to induce nonzero pretilt. For the homologous liquid crystal series alkyl-cyanobyphenyl, we found that the threshold rubbing strength is a monotonic function of the number of methylene units. A dual easy axis model is then used to explain the result.

In chapter 3, Fréedericksz transition measurements have been used to determine the quadratical and quartic coefficients associated with the molecules’ tilt with respect to the layer normal in surface-induced smectic layers in the nematic phase above the smectic-A–nematic phase transition temperature. Both the quadratic and quartic coefficients are consistent with the scaling relationship as predicted in theory, and their ratio is approximately constant.

In chapter 4, a Rayleigh-Taylor instability experiment is performed by using a magnetic field gradient to draw down a low density but highly paramagnetic fluid below a more dense fluid in a Hele-Shaw cell. When turning off the magnetic field, the RT instability occurs \textit{in situ} and the growth of the most unstable wavevector is measured as a function of time. The wavelength of the RT instability along with the growth rate was measured as a function of capillary number (which is related to the density difference and interfacial tension between two fluids). A theory for the instability that permits different viscosities for two immiscible fluids was developed by our colleague Pierre Carlès, and good agreement was found with the experimental results. The technique of magnetic levitation promises to broaden significantly the accessible parameter space of gravitational interfacial instability experiments.

Finally, in chapter 5 we introduced a method to control initial conditions for a
Rayleigh-Taylor instability, wherein a dense fluid sits metastably atop a less dense fluid, a configuration that can be stabilized using a magnetic field gradient when one fluid is highly paramagnetic. On switching off the magnetic field, the instability occurs as the dense fluid falls under gravity. By affixing appropriately shaped magnetically-permeable wires to the outside of the cell, arbitrarily-chosen and well-controlled initial perturbations are imposed on the interface. Using this technique, the perturbation amplitudes, growth rates, nonlinear growth coefficients, and terminal velocities are obtained to examine both the linear and nonlinear growth regimes.

References


Chapter 2

Large Polar Pretilt for the Liquid Crystal Homologous Series Alkylcyanobiphenyl (nCB)

In this chapter, a method of rubbing polyimide at a wedge is used to control pretilt. Sufficiently strong rubbing of the polyimide alignment layer SE-1211 (Nissan Chemical Industries, Ltd.) results in a large pretilt of the liquid crystal director from the homeotropic orientation. The threshold rubbing strength required to induce nonzero pretilt is found to be a monotonic function of the number of methylene units in the homologous liquid crystal series alkylcyanobiphenyl. The results are discussed in terms of the dual easy axis model for alignment.

2.1 Introduction

For liquid crystal alignment, there are two basic forms: homeotropic and planar (or homogeneous) in Fig. 2.1. A uniform alignment may be achieved by coating a
substrate with a thin polyimide layer and rubbed unidirectionally [1]. For displays, it is highly desirable to have a small “pretilt” $\theta_0$ of the nematic director with respect to the substrate normal, in order to avoid tilt domains and increase the switching speed. As a matter of fact, a big pretilt angle may be necessary for some applications in which the optical retardation must be controlled passively by the surface instead of the electric field.

Various methods have been attempted to obtain a high pretilt of liquid crystal director from the homeotropic direction, although only a few techniques have achieved the desired results. For instance, Lee and Seo [2] obtained a pretilt angle of $35^\circ$ with respect to the surface normal for a positive dielectric anisotropic liquid crystal by dipping a homeotropically aligning polyimide in a solvent for 30 minutes. Scharf [3] devised a method by printing a homeotropic aligning agent (or silane) in a grating pattern onto a planar aligning substrate (SiO$_x$). In this printing method, the coverage ratio and the period and the direction of the grating determine the polar and azimuthal liquid crystal orientations such that the full range of pretilt ($0^\circ \leq \theta_0 \leq 90^\circ$) is obtained. Another method to obtain a large pretilt involves
rubbing a polyimide that is designed for homeotropic alignment and has a relatively rigid backbone [4–6]. Although it was found counterintuitively that the pretilt angle decreases with increasing rubbing strength \( n_f \) in reference [4], it was shown that a controllable and temporally robust large pretilt angle \( \theta_0 \geq 40^\circ \) can be obtained above a threshold rubbing strength by using the polyamic acid SE-1211 (Nissan Chemical Industries, Ltd.) and the liquid crystal pentylcyanobiphenyl (5CB) [6]. To understand the origin of the pretilt, Hatoh et al. [7] suggested that the long alkyl side chains of the polyimide that bring about the homeotropic alignment are tilted in the same direction as the rubbing direction. It has been shown that the preparation process of baking at especially high temperature and rubbing results in two concomitant easy axes for orientation: one easy axis is approximately but not necessarily exactly perpendicular to the substrate and the other easy axis is approximately parallel to the substrate [8–10]. Andrienko et al. [11, 12] also suggested the concept of the dual easy axes from their work using polarized ultraviolet light to obtain polar pretilt of the director. In this experiment, we measure the pretilt angle as a function of rubbing strength at substrates coated with a homeotropically aligning polyimide for the homologous series of liquid crystals alkylcyanobiphenyl “nCB” \((5 \leq n \leq 8)\). Our central result is that the threshold rubbing strength \( n_f^{th} \) required to induce nonzero pretilt is found to be a monotonic function of the number of methylene units in the homologous liquid crystal tail, which is consistent with the dual easy axis model [8, 10] and the characteristic behavior of liquid crystals at a surfactant-coated substrate.
2.2 Sample Preparation

In the sample preparation we used the polyamic acid SE-1211 which has a chemical structure shown in Fig. 2.2 dissolved approximately 4% by weight in a varnish. The mixture was spin-coated onto indium tin oxide (ITO)-coated glass slides (3.2 × 0.8 cm) at 1800 rpm for 9 s. The slides were prebaked at 80 °C for 30 min, and then were fully baked at 200 °C for 50 min. This baking temperature is warmer than that prescribed by the manufacturer who suggests 180 °C. Overbaking the polyamic acid further imidizes the backbone, thus promoting nearly planar alignment; it also cleaves away a fraction of the side chains, thereby weakening the homeotropic alignment. Cells made from the slides without rubbing were verified to result in homeotropic alignment for 5CB.

Using a rubbing machine that has a cylindrical roller with a radius of \( r = 4 \) cm, the slides were rubbed with a cotton cloth (Yoshikawa Chemical Co., YA-25-C) having fiber density \( \sigma_f = 1040 \) thread cm\(^{-2} \). With increasing rubbing strength, the backbone of the polyimide SE-1211 becomes well-aligned, allowing the liquid
crystal to overcome the homeotropic alignment tendency due to the side chains [8] and adopt a tilted orientation. To examine multiple rubbing strengths, the slides were rubbed gradiently along the x-axis, as shown in the Fig. 2.3, such that one end of the slide was elevated by an angle of $1.58^\circ$ on the bed of the rubbing machine.

The height of the bed was raised until the cotton fibers osculated the lower end of the slide ($x = 0$) with no deformation of the fiber pile; this defined the $n_f = 0$, i.e., the zero rubbing strength, position. The fiber pile at the elevated end of the slide was deformed by approximately $\delta = 0.088$ cm. The slide was translated with a speed of $s = 0.42$ cm s$^{-1}$ under the rubbing cylinder for a total of $N = 6$ times, with the cylindrical roller rotating at a rate of $\nu = 8.33$ rotations per second. The “rubbing strength” $n_f$ is defined as the number of fibers passing a position of unit width [13] and is given by

$$n_f = (2r\delta)^{1/2}2\pi N\nu r\sigma_f/s$$

(2.1)

$n_f$ was continuously varied with $x$ due to changes in the cotton fiber deformation $\delta$ with position.

A liquid crystal cell was created by placing mylar spacers between two slides: one slide unrubbed and the other one gradiently rubbed, and by adjusting for maximum
parallelism. Using interferometric scheme [14], the cell thickness was measured to be \(l = (15.5 \pm 0.9) \, \mu\text{m}\). Four almost identical cells were prepared. Each of these cells was filled with the liquid crystal nCB in the isotropic phase and then cooled into the nematic phase.

### 2.3 Experimental Setup

As shown in Fig. 2.5, light from a He-Ne laser passed consecutively through a light chopper at frequency of \(f = 237\) Hz, a polarizer oriented at 45° with respect to the rubbing direction of the sample, a Babinet-Soleil compensator, through a cylindrical lens of focal length 150 mm, the sample, an analyzer, a second lens that recollimated the beam, and into a photodetector. The detector output, which is proportional to the total light intensity \(I\), was fed into a lock-in amplifier referenced to the frequency of the chopped light between the laser and the polarizer. The cylindrical lens produced an approximately rectangular beam profile at the sample (0.15 \times 1 \, \text{mm in size}), with the smaller dimension being parallel to the rubbing di-
rection. The shape of the beam served two purposes: The narrow dimension limited the range of rubbing strengths $n_f$ probed by the beam, and the wide dimension allowed us to average over a large area for a given $n_f$. This averaging process was useful because, even though local variations in the rubbing strength (for a given nominal value $n_f$) are small, the induced tilt angle $\theta_0$ can be a strong function of $n_f$; $\theta_0$ could vary by up to $\pm 2^\circ$.

2.4 Optical Retardation Measurement

The apparent pretilt angle $\theta_0$ for each cell was obtained by measuring the optical retardation through the cell. In order to measure the retardation as a function of rubbing strength, the cell was mounted on a horizontal translation stage and was translated along the rubbing direction $x$ (which is the same direction as the gradient of the rubbing strength). For each position corresponding to each value of $n_f$ in the cell, the compensator was adjusted to minimize the signal into the lock-in amplifier, and then the measured value of the optical retardation from the compensator was taken to be equal in magnitude to the sample retardation $\alpha$. For instance, Fig. 2.6 showed the retardation as a function of rubbing strength for 5CB at different temperatures.

2.5 Dual Easy Axis Model

In the dual easy axis picture [8–10], the liquid crystal is influenced by two preferred directions: One is approximately vertical and is due to the polymers side
chains, and the other is approximately planar, i.e., parallel to the rubbed polyimide backbone. The liquid crystal adopts an equilibrium pretilt angle $\theta_0$ due to competition from the two easy axes.

In the orthogonal dual easy axis model for which there is a surface free energy \cite{8}

$$F_{\text{surf}} = A\sin^2 \theta_0 + B\cos^2 \theta_0 + C\sin^4 \theta_0,$$

when $B > A$, a nonzero tilt is obtained as

$$\theta_0 = \sin^{-1} \left[ \frac{(B - A)}{2C} \right]^{\frac{1}{2}}. \quad (2.3)$$

Here $A$ is the quadratic rubbing strength-dependent anchoring strength coefficient for homeotropic alignment \cite{15}, $B$ is the quadratic coefficient for planar alignment \cite{15}, and $C$ is the quartic coefficient required for stability \cite{16}. But the propensity for homeotropic order, and thus $A$, is related to the length of the liquid crystal tail \cite{17}.

Figure 2.6: Retardation as a function of rubbing strength at different temperatures ($\ast$: $T = 25^\circ$; $\square$: $T = 32^\circ$).
2.6 Pretilt Angle

The pretilt angle was determined from the measured retardation as follows. First, sufficiently rigid anchoring conditions were assumed, which was verified previously for $\theta_0 \geq 15^\circ$ [6]. In the dual easy axis picture [8–10], the liquid crystal adopts an equilibrium pretilt angle $\theta_0$ due to competition from the two easy axes. For weak rubbing the side chains dominate and $\theta_0 = 0$. With increasing rubbing strength the influence of the planar easy axis increases, with a corresponding decrease in the polar anchoring strength coefficient. At a threshold rubbing strength $n_{th}^f$, the planar easy axis becomes sufficiently influential that the effective polar anchoring strength vanishes; for $n_f > n_{th}^f$, the pretilt angle becomes nonzero, but with an increasing anchoring strength that is proportional to $\theta_0^2$ [10].

Returning to the experiment at hand, for very small $\theta_0 (\theta_0 \ll 15^\circ)$ the weakened anchoring strength coupled with elastic torque gives rise to an artificially larger measured tilt at the rubbed SE-1211 substrate, i.e., the measured value of $\theta_0$ is larger than the pretilt angle of an isolated substrate. In consequence, the actual retardation vs $n_f$ should be slightly more rounded in the vicinity of $n_{th}^f$. This was not deemed a significant issue for this work, which focuses on $n_{th}^f$ vs homolog number.

The free energy of the deformation is given as follows

$$F_d = \frac{1}{2} \left[ K_{11} \sin^2 \theta(z) + K_{33} \cos^2 \theta(z) \right] \left[ \frac{\partial \theta(z)}{\partial z} \right]^2,$$

(2.4)

where $K_{11}$ is the splay elastic constant and $K_{33}$ is the bend elastic constant. Values of the parameters $K_{11}$ and $K_{33}$ are temperature-dependent and were taken from
Ref. [18] for the calculation. By minimizing the free energy, we obtained the director profile across the cell \( \theta(z) \), which satisfies the following equation:

\[
\left[ K_{11} \sin^2 \theta(z) + K_{33} \cos^2 \theta(z) \right] \frac{\partial^2 \theta(z)}{\partial z^2} = (K_{33} - K_{11}) \sin \theta(z) \cos \theta(z) \left[ \frac{\partial \theta(z)}{\partial z} \right]^2. \quad (2.5)
\]

We then calculated values of the retardation \( \alpha_{\text{calc}} \), assuming a particular value of the pretilt angle \( \theta_0 \) in the relation

\[
\alpha_{\text{calc}} = \int \frac{2\pi}{\lambda} \left[ n_{e}^{\text{eff}}(z) - n_o \right] dz, \quad (2.6)
\]

where \( \lambda \) is the wavelength of light, \( n_o \) is the ordinary refractive index, and \( n_{e}^{\text{eff}}(z) \) is the effective refractive index at the position \( z \) along the normal from the surface of the cell. \( n_{e}^{\text{eff}}(z) \) was obtained from the following relation:

\[
\frac{1}{[n_{e}^{\text{eff}}(z)]^2} = \frac{\cos^2 \theta(z)}{n_o^2} + \frac{\sin^2 \theta(z)}{n_e^2}, \quad (2.7)
\]

where \( n_o \) and \( n_e \) for each homolog at the appropriate temperatures were obtained from Ref. [19], and \( \theta(z) \) is the calculated polar angle of the director at position \( z \).

Finally, the pretilt angle \( \theta_0 \) was adjusted so that \( \alpha_{\text{calc}} \) was matched to the measured value of the retardation \( \alpha \).

Fig. 2.7 shows \( \theta_0 \) vs \( n_f \). It had earlier been shown that these values are temporally robust for the liquid crystal 5CB [6].
Figure 2.7: $\theta_0$ vs $n_f$: Circles correspond to 5CB (open circles at 25°C and solid circles at 32°C), stars to 6CB, squares to 7CB, and triangles to 8CB [upward triangles at 33.75°C (about 0.25°C above $T_{NA}$), rightward triangles at 34°C, leftward triangles at 36°C, and downward triangles at 38°C]. Typical horizontal error bars are shown, and are due primarily to the uncertainty in locating $n_f = 0$. Uncertainty in $\theta_0 \sim \pm 2^\circ$, primarily from the spread in $\theta_0$ due to small variations in rubbing strength.
2.7 Result and Discussion

The pretilt angles at low rubbing strengths (up to \( n_f^{th} \sim 2 \times 10^6 \text{ cm}^{-1} \)) are close to zero. For rubbing strengths \( n_f \) larger than a liquid crystal-dependent threshold value \( n_f^{th} \), the retardation increases continuously to a high value, corresponding to a rise in the pretilt angle from almost zero to approximately 35°. We remark that 35° is not an asymptotic value for the pretilt, and considerably larger pretilts may be obtained with stronger rubbing or more intense baking conditions for the polyamic acid (hotter and/or longer in duration [20]). The existence of a threshold rubbing strength \( n_f^{th} \) below which the pretilt is small or zero is due to a convergence of several phenomena. As discussed in Ref. [8], threshold behavior is a consequence of the existence of two orthogonal easy axes, at least for bidirectional rubbing. Nevertheless, on examining the surface-induced director tilt in the smectic-A phase above the smectic-C phase transition for a unidirectionally rubbed substrate, Syed et al. [9] concluded that the two easy axes need not be mutually orthogonal. This raises a question: If the easy axes are not orthogonal, why is there a threshold rubbing strength to achieve a nonzero pretilt \( \theta_0 \) (rather than a continuous variation of pretilt) in the nematic phase for unidirectional rubbing? We believe the key to this problem is the demonstrated existence [21–23] of a minimum rubbing strength \( n_f^{Y} \) required for disentanglement and reorientation of the polyimide backbone, an effect similar to the yield stress associated with a Bingham fluid [24]. The polyimide cannot become disentangled for \( n_f < n_f^{Y} \), and thus the planar easy axis remains weak. In consequence \( \theta_0 \) remains close to zero. For \( n_f > n_f^{Y} \) the polymer can begin
to disentangle and reorient (parallel to the rubbing direction), thereby increasing the influence of the planar easy axis. The result is a small torque on the director if the easy axes are not orthogonal. This would result in a small pretilt angle $\theta_0$, which increases with increasing rubbing strength \[9\]. For sufficiently large $n_f$ a rubbing strength is reached at which the two easy axes become comparable in strength. This corresponds to $n_f = n_f^{th}$ (which must be larger than $n_f^Y$), and for $n_f > n_f^{th}$ the tilt grows rapidly with increasing rubbing strength \[8\]. In principle a well defined threshold $n_f^{th}$ exists only when the easy axes are perfectly orthogonal, although if the easy axes are nearly orthogonal an approximate threshold rubbing strength $n_f^{th}$ may be identified and $\theta_0$ vs $n_f$ will be slightly rounded in this region.

The significant new feature in this work is the dependence of $n_f^{th}$ on homolog number: $n_f^{th}$ increases with increasing alkyl chain length. A single but tilted easy axis is not likely to engender this behavior, although a pair of (nearly) orthogonal easy axes will. It is well known that surface mono-layers consisting of molecules with longer aliphatic tails enhance the homeotropic ordering of liquid crystals, both from the standpoint of more complete wetting of a nematic layer in the isotropic phase and polar anchoring strength within the nematic phase \[25-30\]. Perhaps more relevant is the observation that for glass coated with the silane surfactant dimethyloctadecylaminopropyl trimethoxysily chloride (DMOAP), Chen et al. observed more complete wetting of longer chain homologs of nCB as compared to the shorter homologs \[17\], indicating the importance of the liquid crystal’s alkyl chain length in promoting homeotropic order. Now consider our alignment layer SE-1211, which
consists of long alkyl side chains that promote homeotropic order and a polyimide backbone that, when aligned, tends to promote planar order. According to the orthogonal dual easy axis model, for longer homologs the rubbing must be stronger to overcome the enhanced homeotropic tendency, i.e., stronger rubbing is required for $B$ to become larger than $A$. Thus, $n_f^{th}$ increases with increasing liquid crystal tail length.

Let me comment on two additional points. First, the shapes of the $\theta_0$ vs $n_f$ curves appear to be similar, although we must exercise caution. As noted above, the response of the polyimide is highly nonlinear with rubbing strength, and therefore it is not obvious that $\theta_0$ should be proportional to the quantity $n_f - n_f^{th}$, where $n_f^{th}$ depends on homolog number. Second, in Ref. [8] Shioda et al. observed that for a given rubbing strength and for a liquid crystal possessing a smectic-A to nematic phase transition, a tilt transition from $\theta_0 \neq 0$ to $\theta_0 = 0$ occurs in the nematic phase on cooling, and that the temperature at which this occurs is lower (closer to $T_{NA}$) for stronger rubbing. The origin of this effect is surface-induced smectic ordering, which favors the homeotropic orientation. Thus, a more strongly rubbed substrate would need to be closer to $T_{NA}$, where the homeotropic tendency is greater, for this tilt transition (i.e, tilt to homeotropic alignment) to occur. Our results for 8CB qualitatively are consistent with this behavior, although perhaps not as convincing as the results in Ref. [8] and Ref. [31]. This is due to the larger surface roughness in the present samples (we found topographical features of 18 nm over distances of 0.8 $\mu$m) which tends to suppress surface-induced smectic order [32].
2.8 Conclusion

Sufficiently strong rubbing of the polyimide alignment layer SE-1211 results in a large pretilt of the liquid crystal director from the homeotropic orientation. The threshold rubbing strength for the onset of nonzero pretilt has been found to increase with an increase in the number of methylene units in the liquid crystal tail, which is consistent with the dual easy axis model and the characteristic behavior of liquid crystals at a surfactant-coated substrate.

References


Chapter 3

Smectic Tilt Susceptibility: Anharmonic Behavior in Surface-induced Smectic Layers above the Nematic–Smectic-A Transition Temperature

In this chapter, Fréedericksz transition measurements were used to determine the quartic contribution to the energy associated with molecular tilt relative to the layer normal in the surface-induced smectic layers in the nematic phase above the smectic-A transition temperature $T_{NA}$. Both the quadratic and quartic coefficients are consistent with the scaling relationship $(T - T_{NA})^{-3\nu}$, where $\nu$ is the correlation length exponent, and their ratio was approximately constant.
3.1 Introduction

The smectic-A (Sm-A) phase is characterized by a periodic density wave of amplitude $\psi$ and wave vector $\mathbf{q}_0$ perpendicular to the average molecular orientation (i.e., the “director” $\mathbf{n}$), which we define as being parallel to the $z$ axis [1]. Two types of elastic distortions are associated with smectic layering: Compression of the layers involves an energy per unit volume that, to lowest order, is given by

$$f_{\text{compr}} = \frac{1}{2} B \left( \frac{\partial u}{\partial z} \right)^2,$$

(3.1)

where $B$ is the compression elastic constant and $u$ is the deviation of the smectic layer from its undisturbed position. Additionally, there is an energy cost per unit volume when the molecules tilt by an angle with respect to the layer normal; to lowest order this is given by

$$f_{\text{tilt}} = \frac{1}{2} D_2 \theta^2,$$

(3.2)

where $D_2$ is the tilt elastic constant with the dimensions of surface tension. Both $B$ and $D_2$ scale as $\langle \psi^2 \rangle$ [1], and increase with decreasing temperature below the nematic–Sm-A phase transition temperature $T_{NA}$, which we found to be 79.5°C for the liquid crystal SCE12R. For $T > T_{NA}$ smectic fluctuations occur in the bulk nematic phase. Moreover, for a sufficiently smooth substrate treated for homeotropic orientation, surface-induced smectic layers appear, such that the order parameter $\psi$ is largest at the substrate and decays into the bulk over a temperature-dependent correlation length $\xi$ [2–8]. In a recent paper Syed and Rosenblatt [9] examined the
effect of surface-induced smectic layering in the nematic phase for a substrate treated to produce tilted alignment well above $T_{NA}$. In this system there are two nematic easy axes, one approximately perpendicular and one approximately parallel to the surface, causing the liquid crystal to adopt a temperature-dependent equilibrium surface tilt angle $\theta_s$ determined by the strengths of the two easy axes [10,11]. Carbone and Rosenblatt [12] found that at temperatures far above $T_{NA}$ the molecules tilt by a nonzero angle $\theta_s(T)$. On cooling, however, the energy term $f_{\text{tilt}}$, which promotes a director orientation perpendicular to the surface-induced smectic layers, becomes dominant and results in $\theta_s(T) \rightarrow 0$ at a temperature $T_a(>T_{NA})$; for $T_{NA} < T < T_a$ we found that $\theta_s = 0$, as shown in Fig. 3.1.

Let us now consider the anchoring properties at the liquid crystal/substrate interface in the purely homeotropic temperature region, viz., $T_{NA} < T < T_a$. In the absence of smectic order, the anchoring energy per unit area is given by

$$F_{\text{anch}} = \frac{1}{2} W_2 \theta_s^2 + \frac{1}{4} W_4 \theta_s^4 + \cdots, \quad (3.3)$$
where $W_2$ and $W_4$ are the quadratic (Rapini-Papoular) [13] and quartic anchoring strength coefficients [14], respectively. Note that $W_2 < 0$ for a surface treated so as to induce a nonzero polar pretilt angle $\theta_s$ [10,12]. To this we now must add the areal contribution

$$F_{\text{tilt}} = \frac{1}{2} \int_0^d dz D_2^0 e^{-\frac{2z}{\xi}} \theta_s^2 \approx \frac{1}{4} \xi D_2^0 \theta_s^2$$

(3.4)
due to surface-induced smectic layering [10], which effectively stiffens the homeotropic anchoring and reduces the molecules ability to tilt. Here it is assumed that $\theta_s$ is approximately constant over the length scale $\xi$ and that $D_2(z) = D_2^0 e^{-2z/\xi}$ reflects the rapid decay of the surface-induced smectic order from the interface into the bulk, where $\xi \ll d$ and $d$ is the cell thickness. Thus, the effective surface free energy per unit area becomes

$$F_{\text{eff}}^{\text{surf}} = \frac{1}{2} \left( W_2 + \frac{1}{2} \xi D_2^0 \right) \theta_s^2 + \frac{1}{2} W_4 \theta_s^4.$$  

(3.5)

Close to $T_{NA}$, where both $D_2^0$ and $\xi$ become large, the coefficient $W_2^{\text{eff}}$ ($= W_2 + \frac{1}{2} \xi D_2^0$) is large and positive, and the director is oriented homeotropically. This is consistent with the large quadratic anchoring strength observed within the Sm-A phase by Li and Lavrentovich [15]. With increasing temperature and a concomitant decrease in surface-induced smectic order, $W_2^{\text{eff}}$ decreases and eventually vanishes at $T_a$ for a substrate treated for nonzero pretilt (because $W_2$ is negative), resulting in the previously observed tilt transition from $\theta_s = 0$ to $\theta_s > 0$ for $T > T_a$ [10]. The interfacial behavior in the region $T_{NA} < T < T_a$, which reflects both the inherent nematic easy axes as well as surface-induced smectic layering, may be examined
by means of a Fréedericksz transition technique [1]. In this experiment an applied electric field competes with surface anchoring and bulk elasticity, such that the director undergoes a nonzero tilt above a threshold field $E_{th}^{LC}$. It easily can be shown that [14]

$$\cot \left( \frac{\pi E_{th}^{LC}}{2U} \right) = \frac{\pi K_{33} E_{th}^{LC}}{W_{2}^{eff} Ud},$$

where $K_{33}$ is the bend elastic constant, is the (negative) electric susceptibility anisotropy, and $U = (\pi/d)(K_{33}/\Delta \chi)^{1/2}$. Here $U$ corresponds to the Fréedericksz threshold field in the limit that the quadratic anchoring strength coefficient $W_{2}^{eff} \to \infty$ and/or $d \to \infty$. Note that $E_{th}^{LC}$ does not depend on beyond-quadratic terms in $F_{surf}$. In this chapter I describe an optical transmission measurements around the Fréedericksz transition threshold for temperatures below the tilt transition temperature $T_{a}$. In addition to finding an expected small threshold field $E_{th}$ on heating toward $T_{a}$ due to the vanishing of $W_{2}^{eff}$, we also observe a very slow initial increase in transmitted optical intensity $I$ with increasing field for $E > E_{th}^{LC}$, behavior that permits us to observe and measure the quartic coefficient in $F_{surf}^{eff}$. Although the quartic and higher order terms play an important role at the bulk smectic-A–smectic-C phase transition where $D_{2}$ vanishes, until now it has been inconsequential at temperatures far above that transition. Our central result is that the quartic coefficient of the tilt energy in the surface-induced smectic layers increases rapidly in the nematic phase as the temperature is decreased toward $T_{NA}$. This result indicates that the molecules’ resistance to tilt in the surface-induced layers involves not only the quadratic coefficient $D_{2}$, but depends critically on beyond-quadratic terms.
3.2 Experimental Preparation

3.2.1 Sample Preparation and Material

Electrically conducting indium tin oxide-coated glass slides were cleaned with detergent, acetone, and ethanol, and then spin coated with the polyamic acid SE-1211 with chemical structure shown in Fig. 2.2, for which there exists a pair of easy axes that result in a nonzero tilt of the director far above $T_{NA}$ [7, 12]. The slides were prebaked for 30 min at 80 °C and baked for 50 min at 200 °C, and then rubbed uniformly with a single pass under a rayon cloth (Yoshikawa Chemical Co., YA-20-R, average fiber density was $\sigma_f = 1411 \text{ threadscm}^{-2}$) using an Optron rubbing machine. The “rubbing strength” $n_f$, which is defined in chapter 2, was $n_f \approx 1.27 \times 10^6 \text{ cm}^{-1}$. Two slides, separated by Mylar spacers of nominal thickness 2 μm, were placed together in an antiparallel configuration, cemented, and placed into a temperature controlled oven. The thickness $d$ of the cell gap, defined as the empty region between the polyimide alignment layers, was measured by an interferometric technique and found to be 4.28 ± 0.05 μm. The negative dielectric anisotropy racemic liquid crystal mixture SCE12R (Merck) was introduced into the cell in the isotropic phase shown in Fig. 3.2 and cooled into the nematic phase.
3.2.2 Experimental Setup

Light from a He-Ne laser at wavelength $\lambda = 633$ nm passed through a polarizer, the sample whose rubbing direction was oriented at $45^\circ$ with respect to the polarizer, an analyzer crossed with respect to the polarizer, and into a photodiode detector, whose output was fed into a digital voltmeter. An ac voltage at frequency $\nu = 1000$ Hz was applied across the liquid crystal cell, as shown in Fig. 3.3.
3.3 Results

The amplitude of the applied voltage was increased in steps of $V_{\text{cell}} = 0.075 \, \text{V}$, each voltage having a dwell time of 6 s before the next increase. I remark that the actual voltage difference $V_{\text{LC}}$ applied across the liquid crystal layer was less than the voltage $V_{\text{cell}}$ applied to the cell, viz., $V_{\text{LC}} = V_{\text{cell}}/[1+d_P/(\varepsilon_P \int_0^d \varepsilon^{-1} \, dz)]$, where $\varepsilon$ is the temperature- and $\theta$-dependent dielectric constant of the liquid crystal. The reduced voltage is due to the capacitive voltage drop across the two polyimide layers, having a combined thickness $d_P = 1.90 \, \mu\text{m}$ (as measured using a Dektak II profilometer) and dielectric constant $\varepsilon_P = 2.9$ (as specified by the manufacturer). Note that data will be plotted as a function of $V_{\text{cell}}$, although modeling will be performed with $V_{\text{LC}}$. Measurements of intensity $I$ vs applied voltage were made first at 87 °C, and subsequent measurements were made at decreasing temperatures toward $T_{N,A}$. Fig. 3.4 shows typical data over part of this temperature range.

There are two features of note for the intensity scale used in Fig. 3.4: (i) there appears to be a Fréedericksz transition at moderately high threshold voltages $V_{\text{cell}}^{\text{th}}$ for all temperatures $T < T_a = 85.5 \, ^\circ\text{C}$—this will turn out not to be the case—and (ii) the intensities at $V_{\text{cell}} = 0$ for $T > T_a$ are significantly larger than the intensities measured at $T < T_a$. Fig. 3.5(a) shows the intensity scale magnified by a factor of 200, and Fig. 3.5(b) shows the scale magnified by a factor of 8000. (Note that, although the intensity is expressed in arbitrary units, these units are the same in both Fig. 3.4 and Fig. 3.5. Thus, the intensity scales shown in the figures may be compared directly.) From Fig. 3.5(a) we see that the intensities
Figure 3.4: Intensity vs voltage $V_{cell}$ applied to the cell at selected temperatures of ■ 87, □ 86, ● 85, ○ 84, ▲ 83, △ 82, ▼ 81.

for $T > T_a$ at $V_{cell} = 0$ vary considerably with temperature and that the slopes $dI/dV_{cell} > 0$ at $V_{cell} = 0$. These features are a clear indication that the director has a nonzero pretilt angle $\theta_s$ in the absence of an applied field. On the other hand, for $T < T_a$ it is obvious from Fig. 3.5(b) that the remnant intensity at $V_{cell} = 0$ is much less temperature sensitive and that the slopes $dI/dV_{cell} = 0$ at $V_{cell} = 0$. (The small differences in intensity at $V_{cell} = 0$ can be ascribed to temperature-dependent variations in the strain birefringence in the glass, as well as to variations of the laser intensity over time.) It is obvious that the initial intensity is very small and remains constant with increasing voltage until a temperature-dependent threshold voltage is reached (shown approximately by the arrows), above which $I$ begins to increase very slowly with voltage [cf. Fig. 3.4 and Fig. 3.5(a)]. Thus the actual threshold
Figure 3.5: Intensity vs $V_{cell}$. Scale enlarged by a factor of (a) 200 and (b) 8000 relative to Fig. 3.4. Solid lines show examples of calculated intensity, and arrows show the threshold voltage at several temperatures. Note that $V_{LC}$ was calculated from the applied voltage $V_{cell}$. Symbols correspond to ◦ 84, ▲ 83, △ 82, and ▼ 81°C.
voltages are considerably smaller than the apparent threshold voltages noted in Fig. 3.4 and item (i) above, with the discrepancy being largest at \( T_a \) (where \( W_{2}^{eff} \) is expected to vanish) and decreasing on lowering the temperature toward \( T_{NA} \) (where \( W_{2}^{eff} \) is expected to become large due to surface-induced smectic ordering). Thus the apparent Fréedericksz transition threshold voltages in Fig. 3.4 are artifacts of the extremely weak rise of the intensity for voltages just above the real Fréedericksz threshold, which actually occurs at lower voltages. It turns out that the intensity vs voltage behavior for very small \( W_{2}^{eff} \) depends strongly on the magnitude of the effective quartic coefficient \( W_{4}^{eff} \), as we shall see below.

### 3.4 Theoretical Model

Due to the translational symmetry of the system with the middle plane located at \( z = d/2 \), the polar angle profile \( \theta(z) \) has

\[
\theta \left( \frac{d}{2} + z \right) = \theta \left( \frac{d}{2} - z \right).
\]

The total volumetric free energy density includes contributions from the elastic distortion, the applied electric field and the surface anchoring energy, which is given by

\[
\mathcal{f}_{tot} = \mathcal{f}_{elastic} + \mathcal{f}_{electric} + \mathcal{f}_{surf}. \tag{3.7}
\]

In a single elastic constant approximation (\( K = K_{33} \)), the nematic elastic distortion energy density is given by

\[
\mathcal{f}_{elastic} = \frac{1}{2} K \left( \frac{d\theta}{dz} \right)^2. \tag{3.8}
\]
Since the polar angle $\theta(z)$ is not constant, the electric field contribution to the free energy has to be expressed in terms of the electric displacement $D$ (related to the applied voltage $V_{LC}$), which is given by

$$f_{electric} = \frac{D^2}{2\varepsilon_\perp(1 - R \cos^2 \theta)}, \quad \text{(3.9)}$$

where $R = (\varepsilon_\perp - \varepsilon_\parallel)/\varepsilon_\perp$, with $\varepsilon_\perp$ and $\varepsilon_\parallel$ being the liquid crystal dielectric constants perpendicular and parallel to the director $\hat{n}$, respectively, and $D$ the magnitude of the electric displacement vector given by

$$D = -\frac{\varepsilon_\perp V_{LC}}{\int_0^d \frac{dz}{(1 - R \cos^2 \theta)}}, \quad \text{(3.10)}$$

The surface anchoring energy is given by

$$f_{surf} = F_{surf}^{eff}[\delta(z) + \delta(z - d)], \quad \text{(3.11)}$$

The Euler-Lagrange equation for the system is

$$K \frac{d^2 \theta}{dz^2} = -\frac{D^2 R \sin \theta \cos \theta}{\varepsilon_\perp(1 - R \cos^2 \theta)}, \quad \text{(3.12)}$$

or equivalently

$$K \left( \frac{d \theta}{dz} \right)^2 - \frac{D^2}{\varepsilon_\perp(1 - R \cos^2 \theta)} = \text{constant}. \quad \text{(3.13)}$$

The constant can be eliminated by using $d \theta/dz = 0$ and $\theta = \theta_m$ in the middle plane $z = d/2$. Thus, Eq. (3.13) can be rewritten as follow
\[ K \left( \frac{d\theta}{dz} \right)^2 = \frac{\cos^2\theta_m - \cos^2\theta}{\varepsilon(1 - R\cos^2\theta)(1 - R\cos^2\theta_m)} RD^2. \] (3.14)

Combining Eq. (3.14) with the torque balance equation given by

\[ K \left( \frac{d\theta}{dz} \right)_{z=0} = W_{2}^{eff}\theta_s + W_{4}^{eff}\theta_s^3, \] (3.15)

we obtain

\[ D = \frac{W_2^{eff}\theta_s + W_4^{eff}\theta_s^3}{K} \sqrt{\frac{K\varepsilon(1 - R\cos^2\theta_s)(1 - R\cos^2\theta_m)}{R(\cos^2\theta_s - \cos^2\theta_m)}}. \] (3.16)

Also, from Eq. (3.14) we obtain

\[ D = \frac{d}{2} \int_{\theta_s}^{\theta_m} d\theta \sqrt{\frac{1 - R\cos^2\theta_s}{\cos^2\theta_s - \cos^2\theta_m}}. \] (3.17)

Combining Eq. (3.16) and Eq. (3.17), we have

\[ \frac{W_2^{eff}\theta_s + W_4^{eff}\theta_s^3}{K} \sqrt{\frac{1 - R\cos^2\theta_s}{\cos^2\theta_s - \cos^2\theta_m}} = \frac{d}{2} \int_{\theta_s}^{\theta_m} d\theta \sqrt{\frac{1 - R\cos^2\theta}{\cos^2\theta_s - \cos^2\theta_m}}. \] (3.18)

From Eq. (3.18) we are able to obtain \( \theta_m \) as a function of the pretilt angle \( \theta_s \). Thus, the electric displacement \( D \) from Eq. (3.16) is obtained as a function of \( \theta_s \).

For a given value of \( \theta_s \) we are able to obtain the polar angle profile \( \theta(z) \) by solving numerically the differential Eq. (3.12) with the boundary conditions:

- \( \theta(z = 0) = \theta_s \).
\[
\left( \frac{d\theta}{dz} \right)_{z=d/2} = 0.
\]

The retardation \( \alpha_{calc} \) can be obtained through the integral of the profile \( \theta(z) \) as follows:

\[
\alpha_{calc} = \frac{2\pi n_\perp}{\lambda} \int_0^d dz \left( \frac{1}{\sqrt{1 - \left(\frac{n_\perp}{n_\parallel}\right)^2 \sin^2 \theta}} - 1 \right),
\]

(3.19)

where \( n_\perp \) and \( n_\parallel \) are the ordinary and extraordinary refractive indices of the liquid crystal, respectively.

### 3.5 Data Analysis

I analyzed the data first by estimating a value for \( T_{NA} \) and obtaining \( W_2^{eff}(T) \) directly from the Fréedericksz threshold voltages \( V_{th} \) applied across the liquid crystal—\( V_{th} \) may be calculated from \( V_{th}^{cell} \) across the cell, as noted above and from Eq. (3.6). \( V_{th}^{cell} \) vs \( T - T_{NA} \) is shown in Fig. 3.6. I then considered the total volumetric free energy density in Eq. (3.7). Because fittings are performed from \( V_{LC} = V_{cell} = 0 \) to a voltage only slightly above \( V_{th}^{LC} \), bend deformation dominates and therefore I used the single elastic constant approximation with \( K = K_{33} \). The dielectric constants \( \varepsilon_\perp \) and \( \varepsilon_\parallel \) were measured using an Andeen-Hagerling 2500 capacitance bridge operating at 1000 Hz; results are shown in Fig. 3.7.

The bend elastic constant \( K_{33} \) was obtained by measuring the Fréedericksz transition threshold for a thick cell (58.6 \( \mu m \))—this threshold voltage corresponds approximately to \( U \) in the thick cell regime—and by taking \( \Delta \chi = (\varepsilon_\parallel - \varepsilon_\perp) / 4\pi \); results for \( K_{33} \) are shown in Fig. 3.8. Using the measured material parameters, exploiting
Figure 3.6: \( V_{\text{cell}}^{th} \) vs reduced temperature \( T - T_{\text{NA}} \). Note that the threshold vanishes above \( T = T_a \), corresponding to approximately 85.5 °C.

Figure 3.7: Dielectric constants \( \varepsilon_{\perp} \) and \( \varepsilon_{\parallel} \) vs reduced temperature \( T - T_{\text{NA}} \).
Figure 3.8: Bend elastic constant $K_{33}$ vs reduced temperature $T - T_{NA}$.

the symmetry of $\theta$ about $z = d/2$, and temporarily estimating a value for the adjustable parameter $W_4^{\text{eff}}$, these equations were solved in a manner similar to that in Ref. [12] to obtain $\theta(z, V_{LC}, T)$. The optical phase retardation [12] $\alpha_{\text{calc}}(V_{LC}, T)$ in Eq. (3.19) then was calculated as a function of temperature using $\theta(z, V_{LC}, T)$. Here $n_\perp = 1.4805$ and $n_{||} = 1.635$ are the ordinary and extraordinary refractive indices, respectively, of the liquid crystal measured at $\lambda = 633$ nm using an Abbé refractometer. Note that these values are nearly independent of temperature over the relevant temperature range. Finally,

$$I_{\text{calc}} = I_0 \sin^2 \left( \frac{\alpha_{\text{calc}}}{2} \right)$$

was calculated for each temperature as a function of $V_{LC}$ (and therefore as a function of $V_{cell}$), where $I_0$ is the maximum intensity (for $\alpha = \pi$), which may be obtained
experimentally from Fig. 3.4. Since I chose an adjustable value of $W^\text{eff}_4$, I then had to examine a sizable region of parameter space for $W^\text{eff}_4$ and $T_{NA}$, where it was necessary to recalculate $W^\text{eff}_2(T)$ for each value of $T_{NA}$. The best agreement between the calculated and experimental intensities corresponds to a fitted value of $T_{NA} = 79.5 \pm 0.2 \, ^\circ\text{C}$, and is shown as a function of $V_{\text{cell}}$ in Fig. 3.5(b) for several temperatures. Values for $W^\text{eff}_2(T)$ and $W^\text{eff}_4(T)$ are shown in Fig. 3.9. Finally, we checked the cell thickness $d$ by applying a voltage $V_{\text{cell}}$ up to 100 V and measuring the nearly saturated retardation. Using this technique, $d$ was identical to the value obtained by interferometry of the empty cell.

Figure 3.9: $W^\text{eff}_2$ (solid circles, left-hand scale) and $W^\text{eff}_4$ (open circles, right-hand scale) vs reduced temperature $T - T_{NA}$. Solid line is a fit of $W^\text{eff}_2$ to the form $W_2 + A[(T - T_{NA})/T_{NA}]^{-3\nu}$ (see text).
3.6 Discussion

The calculated intensities were found to be in good agreement with the experimental results, certainly up to \( I \sim 0.05I_0 \), reflecting the small Fréedericksz threshold, the slow rise of intensity above the threshold, and the more rapid increase of intensity at higher voltages. However, at still higher voltages, where \( I \sim 0.1I_0 \), the experimental values of retardation rise a little more slowly than the theoretical values, requiring a larger voltage to reach \( I_0 \). Because we effectively truncated \( F_{\text{surf}}^{\text{eff}} \) at the quartic term, we believe that this slower increase of the experimental intensity with voltage is due to the presence of sixth—and possibly higher—order terms in the surface free energy density, terms not included in our model. This would imply that \( f_{\text{tilt}} \) requires not only a fourth order term, but higher order terms as well. In other words, tilt with respect to the smectic layer normal is much stiffer than the usual harmonic approximation for \( f_{\text{surf}} \) would suggest. The quartic and higher order terms normally are not observable in light scattering experiments due to the relatively small tilt angle (soft mode) fluctuations in the smectic layers far from the Sm-A–Sm-C transition. In this experiment, however, \( \theta_s \) in the surface-induced smectic layers can become large at sufficiently large voltages, and thus the anharmonic terms in \( f_{\text{tilt}} \) become observable.

Let us now turn to the magnitudes of \( W_2^{\text{eff}}(T) \) and \( W_4^{\text{eff}}(T) \). (As an aside, we first note that measurements were not made too close to \( T_{NA} \) because of the growing correlation length, which has the effect of squeezing down the nematic region that can undergo a Fréedericksz transition. This becomes problematical when
\(\xi \gtrsim 0.01d\) [6]. Both \(W_{2}^{\text{eff}}(T)\) and \(W_{4}^{\text{eff}}(T)\) have a “bare” contribution associated with the anchoring properties of the nematic at a substrate, as well as a contribution due to surface-induced smectic order. Previously Shioda \textit{et al.} [10] had estimated the contribution to \(W_{2}^{\text{eff}}(T)\) from the positive \(\xi D_{0}^{2}\) term: Approximately 1 °C above \(T_{NA}\) we had found \(D_{0}^{2} \approx 3 \times 10^{4}\) erg cm\(^{-3}\) and \(\xi \approx 4 \times 10^{-6}\) cm, resulting in a value for \(\xi D_{0}^{2}\) of order 0.1 erg cm\(^{-2}\). This contribution, which scales as \([(T - T_{NA})/T_{NA}]^{-3\nu}\), where \(\nu\) is the critical exponent for \(\xi\) [10], should be considerably smaller at the higher temperatures examined in this experiment. The data in Fig. 3.9 (closed circles) are consistent with this estimate for \(\xi D_{0}^{2}\), although smaller by a factor of about 3. It is important to bear in mind that \(\langle \psi^2 \rangle\), and thus \(D_{0}^{2}\), depends on the roughness of the particular surface, and may be expected to vary from sample to sample. Another feature is that \(W_{2}^{\text{eff}}(T)\) vanishes at \(T_{a}\), and becomes negative for \(T > T_{a}\) where the surface has been treated so that the ordinary nematic anchoring coefficient \(W_{2} < 0\) [10,12]. If we fit \(W_{2}^{\text{eff}}(T)\) to the form \(W_{2} + A[(T - T_{NA})/T_{NA}]^{-3\nu}\), keeping \(\nu = 0.7\) [10] and \(T_{NA} = 79.5\) °C, and allowing \(A\) and \(W_{2}\) to vary, we find that \(W_{2} = -0.0013\) erg cm\(^{-2}\), a very reasonable value in the temperature region where surface-induced smectic order can be ignored and the director has a nonzero polar tilt \(\theta_s\) at \(V_{cell} = 0\). The fit is shown in Fig. 3.9, although it must be kept in mind that precision is limited due to experimental error. Values for \(W_{4}^{\text{eff}}(T)\) (open circles, right-hand scale, Fig. 3.9) are larger than \(W_{2}^{\text{eff}}(T)\).

Previously Carbone and Rosenblatt [12] measured the ordinary contribution \(W_{4}\) for the liquid crystal pentylycyanibiphenyl (5CB), which does not have a Sm-A phase,
finding $W_4 \sim 1 \text{ erg cm}^{-2}$. The presence of surface-induced smectic order increases $W_4^{\text{eff}}(T)$, although far above $T_{NA}$ the smectic contribution to $W_4^{\text{eff}}(T)$ is small, and thus $W_4^{\text{eff}}(T)$, as expected, is comparable to the previous result for $W_4$ for 5CB [12]. It also is interesting to note that the ratio $W_2^{\text{eff}}(T)/W_4^{\text{eff}}(T)$ does not vary systematically with temperature, indicating that the dependence on $\langle \psi^2 \rangle$ and $\xi$ is similar for both exponents. Thus $W_2^{\text{eff}}(T)$ dominates $f_{\text{surf}}$ for small $\theta_s$, but for $\theta_s \gtrsim 10^{-2}$ rad the quartic term becomes larger.

### 3.7 Summary

To summarize, we have demonstrated the existence of and measured as a function of temperature the leading anharmonic term in the inverse tilt susceptibility for surface-induced smectic order in the nematic phase, and shown that it has a significant effect on the director profile above the Fréedericksz transition threshold voltage.

### References


Chapter 4

Rayleigh-Taylor Instability for Immiscible Fluids of Arbitrary Viscosities: Magnetic Levitation Investigation and Theoretical Model

In this chapter, I describe an experiment in which a magnetic field gradient was used to draw down a low density but highly paramagnetic fluid below a more dense fluid in a Hele-Shaw cell. On turning off the field a Rayleigh-Taylor instability was observed in situ, and the growth of the most unstable wavevector was measured as a function of time. A theory for the instability that permits different viscosities for two immiscible fluids was developed, and good agreement was found with the experimental results. The technique of magnetic levitation promises to broaden significantly the accessible parameter space of gravitational interfacial instability experiments.
4.1 Introduction

Interfacial gravity-driven instabilities, including the Rayleigh-Taylor (\textit{RT}) \cite{1,2} and Richtmyer-Meshkov \cite{3–6} instabilities, represent a lively field of fundamental research having myriad applications ranging from stellar physics to inertial fusion to inkjet printing. In an RT experiment, one needs to create two or more layers of immiscible fluids having different densities, and then subject the system to a destabilizing body force. Since gravity always is present, careful control of the initial conditions generally is not possible in the unstable case, where the upper fluid is denser than the lower fluid: the fluid layers destabilize even before they are fully created. In order to circumvent this difficulty, a solid partition may be used to separate the two fluids initially. The partition then is removed suddenly, allowing the instability to occur \cite{7,8}. Unfortunately, this procedure tends to generate unknown and uncontrollable disturbances, which confound interpretation of the experimental results. Another approach involves an initially stable configuration (denser fluid at the bottom), for which two protocols have been used: (i) rapidly turning the cell upside down at $t = 0$ \cite{9} and (ii) imposing a linear vertical acceleration using a spring-loaded mechanism \cite{10–12}. Both techniques have obvious limitations: the first is limited to very slow instability processes, usually obtained through the use of highly viscous and/or immiscible fluids, and the second requires large installations, is time-limited, and does not facilitate temporal control of the total body force after the cell is accelerated.

Here we report on the use of our technique of time varying magnetic levitation
(ML) of fluids [13,14] to study the RT instability in a Hele-Shaw cell using immiscible fluids. We present a new theoretical model developed by our colleague Pierre Carlès of University of Paris 6, constructed for fluids of arbitrary properties, and compare its predictions with experimental data obtained with ML in a parameter regime where other experimental techniques would be difficult, if not impossible, to apply. The very good agreement demonstrates an important and general result concerning this class of instabilities in Hele-Shaw cells. Beyond its inherent interest in terms of fluid dynamics, this result serves as a first example of how ML can dramatically improve our knowledge of gravity-driven interfacial instabilities.

Although the $RT$ instability in magnetic fluids has been studied experimentally in the past [15], it was only in the context of ferromagnetic fluids, which are very different from classical fluids. Here we use ML of classical fluids to stabilize artificially an otherwise unstable fluid configuration (denser fluid on top) for time $t < 0$, allowing us to impose carefully controlled initial conditions. Then at $t = 0$ the magnetic force is turned off rapidly, thereby allowing gravity to destabilize the system and drive the denser fluid to the bottom. In this way we avoid the limitations of previous methods. For the case reported herein we examine two immiscible fluids in a Hele-Shaw cell, a problem that has received scant attention compared to the case of miscible fluids [16,17] because of the rapid destabilization of the interface, rendering the previous techniques impractical. Owing to this paucity of experimental data, theoretical analysis for immiscible fluids also have been scarce and limited to highly idealized situations [16]. Thus, in tandem with our experimental measure-
ments on immiscible fluids using ML, we have developed a new theoretical model for the \( RT \) instability in Hele-Shaw cells that is applicable to immiscible viscous fluids of arbitrary properties. Very good agreement between theory and experiment was found: in particular, we establish that two fluids of different viscosities behave \textit{almost} like fluids with the same viscosity, equal to the average of the two. Thus, both the experimental technique and new theory open a broad range of heretofore inaccessible parameter space.

### 4.2 Carlès’ Theoretical Model

For immiscible fluids in a Hele-Shaw cell subjected to the RT instability in Fig. 4.1, the only extant model is that of Fernandez et al. [17], applicable only to a pair of fluids of the same viscosity—a rather difficult condition to fulfill with experimental
constraints on their mutual miscibility, miscibility of a paramagnetic dopant, and densities. We therefore developed a far more general model that is applicable to our experiment. Our model is based on a fluid cell of infinite extension in the vertical plane \((O; \bar{x}; \bar{z})\) (with \(\bar{x}\) horizontal and \(\bar{z}\) vertical) and of thickness \(h\), corresponding to the \(\bar{y}\) axis. From now on, dimensional space and time variables are indicated by a bar, while their dimensionless counterparts have no bar. Two immiscible fluids of different properties coexist in the cell, separated by an initially horizontal interface and subjected to a gravitational acceleration \(g\) along the \(\mathbf{-\bar{z}}\) direction. We designate the top fluid as 1 and the bottom fluid as 2. The fluids have densities \(\rho_1\) and \(\rho_2\), viscosities \(\eta_1\) and \(\eta_2\), and an interfacial tension \(\gamma\). \(g\) stands for the gravitational acceleration along the \(\mathbf{-\bar{z}}\) direction. We use the Brinkman equations [18], which are equivalent to Navier-Stokes equations averaged over the thickness of the fluid cell (along \(\bar{y}\)). This averaging assumes that the velocity profile in the \(\bar{y}\) direction is Poiseuille-like, which imposes conditions on the typical time of the system evolution, viz. on this typical time scale the vorticity must already have diffused across the cells thickness. The system is made dimensionless by the following choice of variables:

\[
\bar{u}_{ref} = \frac{(\rho_1 - \rho_2)gh^2}{6(\eta_1 + \eta_2)},
\]

\[
\bar{t}_{ref} = \frac{h}{\bar{u}_{ref}},
\]

\[
t = \frac{\bar{t}}{\bar{t}_{ref}},
\]

\[
(x, z) = \left(\frac{\bar{x}}{h}, \frac{\bar{z}}{h}\right),
\]
\[ (u, w) = \left( \frac{\tilde{u}}{\tilde{u}_{ref}}, \frac{\tilde{w}}{\tilde{u}_{ref}} \right), \]

\[ P = \frac{2\tilde{P}}{[(\rho_1 + \rho_2)\tilde{u}_{ref}^2]}. \]

Here \( u \) and \( w \) are the dimensionless velocities along the dimensionless axes \( x \) and \( z \), respectively (with \( \tilde{u} \) and \( \tilde{w} \) their dimensional counterparts), and \( P \) the dimensionless pressure (with \( \tilde{P} \) its dimensional counterpart).

With these definitions the dimensionless equations for each fluid \( i \) become:

\[ 0 = \frac{\partial u_i}{\partial x} + \frac{\partial w_i}{\partial z}, \quad (4.1) \]

\[ 0 = -\frac{\partial P_i}{\partial x} + \frac{1}{Re_i} \left( -12u_i + \frac{\partial^2 u_i}{\partial x^2} + \frac{\partial^2 u_i}{\partial z^2} \right), \quad (4.2) \]

\[ 0 = -\frac{\partial P_i}{\partial z} + \frac{1}{Re_i} \left( -12w_i + \frac{\partial^2 w_i}{\partial x^2} + \frac{\partial^2 w_i}{\partial z^2} \right) - \frac{1}{Fr_i^2}, \quad (4.3) \]

with Reynolds number \( Re_i = (\rho_1 + \rho_2)\tilde{u}_{ref}h/2\eta_i \) and Froud number \( Fr_i^2 = (\rho_1 + \rho_2)\tilde{u}_{ref}^2/2\rho_i gh \). The interface equation is \( z = \zeta(x, t) \).

Far from the interface the boundary conditions are \( \lim_{z \to +\infty} \partial P_1/\partial z = -1/Fr_1^2 \) and \( \lim_{z \to -\infty} \partial P_2/\partial z = -1/Fr_2^2 \), and at the interface itself (i.e., for \( z = \zeta \)):

\[ u_1 = u_2, \]

\[ w_1 = w_2, \]

\[ \frac{\partial \zeta}{\partial t} = w_i - u_i \frac{\partial \zeta}{\partial x}, \]

\[ \frac{\partial u_1}{\partial z} + \frac{\partial w_1}{\partial x} = \frac{\eta_2}{\eta_1} \left( \frac{\partial u_2}{\partial z} + \frac{\partial w_2}{\partial x} \right), \]
\[ P_2 - \frac{2}{Re_2} \frac{\partial w_2}{\partial z} = P_1 - \frac{2}{Re_1} \frac{\partial w_1}{\partial z} - \frac{1}{We} \frac{\partial^2 \zeta}{\partial x^2} \]

where \( We = (\rho_1 + \rho_2) \bar{u}_{r,e}^2 h / 2\gamma \) is the Weber number.

The conditions at the interface represent the continuity of velocity across the interface and the continuity of tangential and normal stresses corrected by the surface tension. If the interface initially is horizontal and perfectly flat between two motionless fluids, then the fluid layers remain in place. In order to check the stability against small interfacial or bulk perturbations, a linear stability analysis was conducted in which a normal mode decomposition was applied to the unknown perturbations using the form \( X = \tilde{X}(z) \exp(ikx + \sigma t) \), where \( X \) is any unknown perturbation of the problem (including the perturbation in \( \zeta \)), \( k \) is the dimensionless wavevector of the perturbation and is real, and \( \sigma \) its dimensionless complex growth rate. Following a classical but lengthy procedure (see e.g., [19]), an explicit dispersion relationship can be found having the form \( \sigma = f(k, We, Re_1, Re_2, Fr_1, Fr_2) \). This relationship can be simplified such that only two dimensionless numbers appear, the capillary number \( Ca = (\eta_1 + \eta_2) \bar{u}_{r,e} / 2\gamma = (\rho_1 - \rho_2) gh^2 / 12\gamma \) and the normalized dynamic viscosity difference \( \mu = (\eta_1 - \eta_2) / (\eta_1 + \eta_2) \), where \(-1 \leq \mu \leq 1\):

\[
\sigma = \frac{(\sqrt{k^2 + 12} - k) \left( k - \frac{k^3}{12Ca} \right)}{2(\sqrt{k^2 + 12} - \mu^2 k) + \mu^2(\sqrt{k^2 + 12} - k) \left[ \frac{3k^2}{15} - \frac{k^3}{18}(\sqrt{k^2 + 12} - k) \right]}. \tag{4.4}
\]

Note that the growth rate is always real in the unstable case, i.e., the interface undergoes no temporal oscillations. If the two viscosities are equal (i.e. \( \eta_1 = \eta_2 \)), we recover Fernandez’s special case result [16]. The present result is thus a general-
ization of that previous theory, applicable to fluids of arbitrary viscosities.

4.3 Hele-Shaw Cell Construction and Two Working Fluids

A Hele-Shaw cell was constructed using glass plates of thickness 0.22 cm and coated with a layer of triethoxysilylpropyl polyethylen oxide urethane to reduce wetting difficulties, as shown in Fig. 4.1. A U-shaped spacer of thickness 0.07 cm was placed between two plates and cemented, creating a fluid “tank” of inner dimensions approximately 6.5 cm in width, 4.5 cm in height, and 0.07 cm in thickness. In order to increase the capillary number, cells with thickness of 0.2 and 0.3 cm, respectively, were constructed using 0.1 cm thick micro slide glass spacer.

The base working fluids were chloroform and water, which are nearly immiscible. Rubrene dye was added to chloroform (1 wt.-\%) in order to create a visual contrast between the two fluids, while highly paramagnetic manganese chloride tetrahydrate (MnCl$_2$·4H$_2$O) was added to the weakly diamagnetic water in varying concentrations, rendering the aqueous solution paramagnetic. The surfactant dodecyltrimethylammonium chloride (DTAC) then was added to some of the extant aqueous mixtures (1 wt.-\% by default) to reduce the surface tension between the fluids and to mitigate wetting problems. For some of the aqueous mixtures glycerol also was added in order to increase the viscosity of the mixtures and thereby achieve larger capillary number while at the same time keeping the system in the Hele-Shaw flow regime, i.e., maintaining a Poiseuille velocity profile across the cells thickness.
4.3.1 Density Measurement

The density of the mixture of chloroform doped with dye was measured to be 1.472 g cm\(^{-3}\), and the density of the aqueous mixtures ranged from 1.316 to 1.394 g cm\(^{-3}\), facilitating a variety of density contrasts between the two fluids. Here is an example how I do the density measurement. A calibrated adjustable pipette (Model: ) with a removable tip was used to measure the mass of a liquid as a function of an apparent volume \(V_{app}\) ml, and then the data were fitted to a linear function of the mass versus the apparent volume. Fig. 4.2 shows the mass of the distilled water as a function of an apparent volume for calibration, since the distilled water has a density 1.0 g cm\(^{-3}\). By replacing the distilled water with chloroform and measuring the mass versus the apparent volume, as shown in Fig 4.3, the density of chloroform was obtained.

\[
\rho_{\text{chloroform}} = \frac{\rho_{\text{chloroform}}}{\rho_{\text{water}}} = \frac{-0.08513 + 1.46879 \times V_{app}}{-0.05024 + 0.98967 \times V_{app}},
\]

For instance, by setting \(V_{app} = 0.95\) ml, \(\rho_{\text{chloroform}} = 1.472\) g cm\(^{-3}\).

4.3.2 Viscosity Measurement

Viscosities of chloroform + dye and the fluid mixtures were measured using Cannon-Ubbelohde viscometers covering the appropriate viscosity ranges, as shown in figure 4.4. For instance, through tube L, the chloroform + dye is filled to the lower reservoir A until its meniscus is between the maximum and the minimum fill lines marked above XX on the left-hand side of U-shaped viscometer, and then
Figure 4.2: Pipette calibration with the distilled water. Apparent volume is the reading on the pipette.

Figure 4.3: Pipette calibration with chloroform.

Figure 4.4: Schematic of a setup for viscosity measurement. The picture of the U-shaped viscometer is taken from the manufacturer [20].
suction from a vacuum pump was applied to tube N until chloroform + dye reached the center of bulb marked D while the tube M was sealed with a finger or stopper. When the suction by the pump and the finger or stopper were removed from the viscometer, the top level of chloroform + dye was allowed to flow freely down toward mark E. The time was measured when the top level of chloroform + dye passed from mark E to mark F. Thus, the kinematic viscosity of chloroform + dye was calculated by multiplying the time by the viscometer constant from the manufacturer. Subsequently, the dynamical viscosity of chloroform + dye was obtained by multiplying the kinematic viscosity of chloroform + dye by its density.

4.3.3 Interfacial Tension Measurement

Interfacial tensions were measured using the pendant drop technique [21]. Fig. 4.5 shows the experimental setup.

In this method, two parameters of the pendant droplet have to be experimentally determined. One parameter is the equatorial diameter $R$ and the other is the diameter $r$ at a distance $R$ from the top of the droplet, as shown in Fig. 4.6 [21].

The interfacial tension is then calculated as follow:

$$\gamma = \frac{\Delta \rho g R^2}{H}$$  \hspace{1cm} (4.5)

with $H$ depending on the “shape factor” $S_f = r/R$. $H$ can be determined by using empirical formula as follow [21, 22]

$$\frac{1}{H} = \frac{T_4}{S_f^4} + T_3 S_f^3 - T_2 S_f^2 + T_1 S_f - T_0$$  \hspace{1cm} (4.6)
Figure 4.5: A photo of the experimental setup for the interfacial tension measurement. A pipette is used to produce a droplet of a heavy fluid in a light fluid. A green laser is used to induce fluorescence of dye in the light fluid for illumination of the interface between two fluids.
Figure 4.6: A pendant droplet for the interfacial tension measurement.

Table 4.1: Empirical constants for the shape dependent parameter $H$ [21, 22].

<table>
<thead>
<tr>
<th>Range of $S_f$</th>
<th>$Q$</th>
<th>$T_1$</th>
<th>$T_2$</th>
<th>$T_3$</th>
<th>$T_4$</th>
<th>$T_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.401-0.460</td>
<td>2.56651</td>
<td>0.32720</td>
<td>0</td>
<td>0.97553</td>
<td>0.84059</td>
<td>0.18069</td>
</tr>
<tr>
<td>0.460-0.590</td>
<td>2.59725</td>
<td>0.31968</td>
<td>0</td>
<td>0.46898</td>
<td>0.50059</td>
<td>0.13261</td>
</tr>
<tr>
<td>0.590-0.680</td>
<td>2.62435</td>
<td>0.31522</td>
<td>0</td>
<td>0.11714</td>
<td>0.15756</td>
<td>0.05285</td>
</tr>
<tr>
<td>0.680-0.900</td>
<td>2.64267</td>
<td>0.31345</td>
<td>0</td>
<td>0.09155</td>
<td>0.14701</td>
<td>0.05877</td>
</tr>
<tr>
<td>0.900-1.000</td>
<td>2.84636</td>
<td>0.30715</td>
<td>-0.69116</td>
<td>-1.08315</td>
<td>-0.18341</td>
<td>0.20970</td>
</tr>
</tbody>
</table>

where $T_i (i = 0, 1, 2, 3, 4)$ and $Q$ are empirical constants for a certain range of $S_f$ and are given in Table 4.1. In order to obtain good quality and reproducible results, it is required to use an extremely clean needle or pipette tip to avoid wetting outer surface of the needle or tip. It is recommended that the diameter of needles is less than half the diameter of the pendant droplet. However, the diameter of the needle should not be too small because the value of $r$ is reduced. As a result, the precision
Table 4.2: Material parameters.

<table>
<thead>
<tr>
<th>MnCl₂·4H₂O (wt. − %)</th>
<th>Density (g cm⁻³)</th>
<th>Dynamic Viscosity (cP)</th>
<th>Interfacial Tension (erg cm⁻²)</th>
<th>Ca</th>
</tr>
</thead>
<tbody>
<tr>
<td>47.0 ± 0.1ᵃ</td>
<td>1.316 ± 0.001</td>
<td>6.20 ± 0.15</td>
<td>0.88 ± 0.1</td>
<td>0.0710</td>
</tr>
<tr>
<td>48.0ᵃ</td>
<td>1.322</td>
<td>6.46</td>
<td>1.02</td>
<td>0.0591</td>
</tr>
<tr>
<td>54.9</td>
<td>1.351</td>
<td>4.31</td>
<td>1.62</td>
<td>0.0300</td>
</tr>
<tr>
<td>56.4</td>
<td>1.371</td>
<td>5.22</td>
<td>2.01</td>
<td>0.0202</td>
</tr>
<tr>
<td>57.9</td>
<td>1.384</td>
<td>6.14</td>
<td>2.28</td>
<td>0.0154</td>
</tr>
<tr>
<td>58.6</td>
<td>1.394</td>
<td>7.11</td>
<td>2.46</td>
<td>0.0128</td>
</tr>
<tr>
<td>30.0ᵇ</td>
<td>1.329</td>
<td>58.42</td>
<td>19.08</td>
<td>0.0245</td>
</tr>
<tr>
<td>30.0ᶜ</td>
<td>1.329</td>
<td>58.42</td>
<td>10.04</td>
<td>0.0465</td>
</tr>
<tr>
<td>30.0ᵈ</td>
<td>1.329</td>
<td>58.42</td>
<td>3.93</td>
<td>0.1187</td>
</tr>
<tr>
<td>30.0ᵉ</td>
<td>1.329</td>
<td>58.42</td>
<td>3.06</td>
<td>0.1526</td>
</tr>
<tr>
<td>30.0ᶠ</td>
<td>1.3297</td>
<td>61.40</td>
<td>2.02</td>
<td>0.2296</td>
</tr>
<tr>
<td>30.0ᵍ</td>
<td>1.3295</td>
<td>59.46</td>
<td>2.09</td>
<td>0.5002</td>
</tr>
</tbody>
</table>

ᵃcontains approximately 9% glycerol and 1% DTAC; ᵇcontains approximately 49% glycerol; ᶜcontains approximately 49% glycerol and 0.1% KCl and 0.05% DTAC; ᵈcontains approximately 49% glycerol and 0.1% KCl and 0.5% DTAC; ᵉcontains approximately 49% glycerol and 0.1% KCl and 0.8% DTAC; ᶠcontains approximately 49% glycerol and 0.2% KCl and 1% DTAC; ᵍcontains approximately 49% glycerol and 0.2% KCl and 1% DTAC.

of the interfacial tension measurement is significantly affected.

Typically, in our experiment r ranges from 20 to 40 pixels and R ranges from 30 to 50 pixels. The calibration of r and R is performed through a spherical metal ball (with a diameter of 5.56 mm) sitting in the bottom of cuvette, as shown in Fig. 4.5.

I remark that I didn’t observe any deformation of the image with the resolution of 1286 pixels × 960 pixels.

4.3.4 Material Parameters for the Aqueous Mixtures

Material parameters for the mixtures are given in Table 4.2.
4.4 Magnetic Levitation

An electromagnet with special Faraday pole pieces was used to produce $\nabla H^2$ uniform to approximately 6% over the length of a 1-cm long column [13, 14, 23], as shown in Fig. 4.7. The magnetic field profile was measured by using a Bell model 9000 Gaussmeter utilizing a Hall effect probe. Fig. 4.8 shows both $H_x$ and the product $H_z \partial H_x / \partial z$ as functions of $z$ when the separation between two pole pieces is 1.7 cm. Note that the $z$-component of field $H_z$ vanishes along the plane $x = 0$, although a small $z$-component of field $H_z$ exists for $x \neq 0$. Over the smaller diameter of the columns $H_z \partial H_x / \partial z$ and $H_z \partial H_z / \partial x$ are small and $H_z \partial H_z / \partial z$ dominates $\nabla H^2$. Therefore, the $z$-component of force is considered to be $\chi H_z \partial H_z / \partial z$ for a material of volumetric magnetic susceptibility $\chi$, and the $x$-component of force $\chi H_z \partial H_z / \partial z$ is small.

4.5 Data Acquisition and Analysis

For all measurements the chloroform + dye mixture, which was treated as a nonmagnetic fluid with susceptibility ($\chi \approx 0$), was denser. Thus, in the absence of an applied magnetic force (per unit volume) $F_{mag} = \chi H \nabla H$, the chloroform + dye sat at the bottom of the cell and the aqueous mixture at the top. The cell was placed in a Faraday magnet whose pole pieces are designed to impart a uniform magnetic force (as opposed to a uniform field), and a downward magnetic force was applied. This created an artificially stable layering of the less dense but paramagnetic aqueous solution at the bottom of the cell and the more dense chloroform + dye at the top.
Figure 4.7: A schematic of magnetic levitation.

Figure 4.8: Magnetic field $H_x$ (right axis) and $H_x \partial H_x / \partial z$ (left axis) as functions of position $z$ when the separation between two pole pieces is 1.7 cm. Note that $z = 0$ corresponds to the position of closest approach of the pole pieces and the quantity $H_x \partial H_x / \partial z$ is maximum at $z = -0.8$ cm.
Figure 4.9: The characteristic decay time of the magnetic field. Note that the response time of the magnetic force is a half of that of the magnetic field (=118 ms).

Once the interface between the two layers stabilized, the current in the magnet was switched off, with $F_{\text{mag}}$ having a characteristic decay time $\tau = 0.06$ s. As shown in Fig. 4.9, the characteristic decay time was measured through the output of LeCroy oscilloscope, which was used to measure simultaneously the voltage across the resistor in the circuit and the output of a digital function generator which controls the power supply to the magnet, as shown in Fig. 4.10. Basically, a $L-R$ circuit is involved in the experimental setup and the time constant is $\tau = L/R$, where $L$ is the inductance and $R$ the resistance in the circuit. We notice that the decay time can be reduced by increasing the resistance in the circuit. As a result, the current in the circuit will be reduced. Since we only need a minimum current to produce a magnetic force sufficiently enough to levitate the fluid 1 (the heavy
Figure 4.10: A schematic of experimental setup for the measurement of the characteristic decay time.

fluid) over the fluid 2 (the light fluid), the decrease of the current will not affect our experimental results. On the other hand, we will avoid a high inductance magnet because we want the decay time as short as possible. Also, the magnetic force goes as $H \nabla H$, which means that the magnetic force relaxation time is half that of the magnetic field. Although we can increase the resistance $R$ to achieve a shorter relaxation time, we have a limitation since we need a bigger power supply to reach the minimal current.

Thereby the fluids were subject to an ordinary gravitational acceleration. With only natural gravity acting on it, the fluid interface became unstable. A digital camera having resolution $768 \times 494$ pixels was used to image the interface deformation at a rate of 30 frames s$^{-1}$ in Fig. 4.11, and a numerical algorithm was applied
Figure 4.11: Images of interfacial region (not the entire cell) at time (a) 0.67 s, (b) 2.00 s, (c) 2.33 s, and (d) 2.67 s after field is turned off. Aqueous solution is 56.4 wt.-% MnCl$_2$·4H$_2$O in water. The interfaces fitted from the digitized images are shown by the dashed lines. Horizontal and vertical scales are the same. Bright spot is a reflection from the light source.

to each frame in order to digitize the interface (see Appendix A for details). The interface was fitted to the four-parameter function $A\sin(2\pi/\lambda + \varphi) + A_0$ (I should remark the fact that the fitting was to a single sinusoid, and did not precisely match the interface shape. But in the analysis, we neglected higher Fourier components, which is a reasonable approximation as long as the amplitude wasn’t too large). Although the wavelength $\lambda$ and phase factor $\varphi$ were difficult to fit for early times, both quickly converged on asymptotic values at later times, as shown in Fig. 4.12 for the interface between the aqueous mixture and chloroform + dye. Note that the offset in Fig. 4.12 decreases as a function of time, which is due to the heavy fluid (chloroform) wetting the edges of the tank. These values then were used to refit the interface for all times with the two adjustable parameters $A$ and $A_0$, as
shown in Fig. 4.13. Thus, for each experimental run we deduced the wavelength of the dominant perturbation and its time evolution, including its linear initial growth rate, over the experimentally accessible material parameters.

### 4.6 Results and Discussions

Let us denote by $\bar{\sigma}$ the dimensional counterpart of $\sigma$, namely, $\bar{\sigma} = \sigma / \bar{t}_{\text{ref}}$. Fig. 4.14 shows the dimensional growth rates $\bar{\sigma}$ measured experimentally and predicted by theory; good agreement with experiment is found over the entire range of Ca. Fig. 4.15 shows the growth rate $\sigma$ and the most unstable wavevector $k$ in dimensionless form, along with the theoretical curves calculated from Eq. (4.4). The two figures show good agreement between experiment and theory. In particular, the
Figure 4.14: Dimensional growth rate $\bar{\sigma}$ for fastest growing perturbation vs Ca. Open triangles represent experiment and solid triangles theory.

Figure 4.15: Dimensionless growth rate $\sigma(=\frac{d(logA)}{dt}t_{ref})$, (open circles) and dimensionless wave vector $k$ (open squares) for fastest growing perturbation, vs Ca. The two theoretical curves for each parameter correspond to the extreme values of $\mu^2$ [0 (solid line) and 1 (dotted line)]. Error bars for $k$ and $\bar{\sigma}$ due to fittings are approximately the size of the data symbols ($\pm 5\%$).
comparison between the theoretical and experimental most unstable wavevectors $k$
is excellent over the entire range of Ca. This agreement confirms both the validity
of the model and the precision of the data acquisition. We stress that the quality
of the experimental results is a direct consequence of the very precise control over
the initial conditions obtained through magnetic levitation. It demonstrates the
great advantage of this technique in the experimental study of Rayleigh-Taylor and
related instabilities.

Another striking observation from the theoretical plots in Fig. 4.15 is the very
small influence of reduced viscosity $\mu$ on the most unstable wavevector. Regardless
of the viscosity difference, the two limiting curves are nearly identical. Nevertheless,
one should not infer that the viscosity differences play no role: The overall shape
of the relationship that links $\sigma$ and $k$ at constant Ca does differ depending on the
value of $\mu$, but the position and amplitude of its maximum remain almost the same,
regardless of $\mu$. Hence the similarity of the curves in the figures. Besides, the di-

censionless growth rate is normalized by $\bar{t}_{ref}^{-1}$, which is a function of the average of
the two viscosities of the fluids, so that even if the dimensionless maximum growth
rate $\sigma$ seems little affected by the viscosity contrast, the dimensional growth rate
$\bar{\sigma}$ is. The main conclusion to draw from this model is that for the most unstable
mode in the linear regime, a layering of immiscible fluids of different viscosities can
be regarded almost as a layering of fluids of equal viscosities, having a value equal
to the arithmetic mean of the two actual viscosities. This prediction is of utmost
importance experimentally, as identifying immiscible fluids of different densities but
equal—or even similar—viscosities generally is not possible. This result shows that even pairs of fluids with high viscosity contrast can be considered a single viscosity system with this explicit value for the viscosity. This conclusion is somewhat reminiscent of earlier results obtained by Menikoff et al. [24] and Mikaelian [25]. In both of these cases an approximate calculation of the dispersion relationship for the Rayleigh-Taylor case showed that to some degree of approximation, the only relevant viscous parameter is the density-averaged viscosity. In this chapter we use an exact and explicit calculation of the dispersion relationship to derive an analogous, although not equivalent, criterion for the Hele-Shaw configuration.

4.7 Summary and Prospect

In summary, magnetic levitation was used to examine gravitational interfacial instabilities. This experimental procedure was shown to be extremely effective in controlling the initial conditions, while at the same time avoiding most of the limitations of more classical methods. Because magnetic levitation facilitates experiments over a wider parameter space, a theoretical model for the Rayleigh-Taylor instability in a Hele-Shaw cell in which the fluids have different viscosities was developed, showing good agreement with experiment. Natural extensions of this work, besides measurements over a broader range of Ca numbers, will be the study of a pure Rayleigh-Taylor regime in thicker cells, and the use of magnetically-created artificial gravity to examine interface instabilities under temporally variable accelerations, such as the Richtmeyer-Meshkov instability [5,6,26].
References


Chapter 5

Rayleigh-Taylor Instability
Experiments with Precise and Arbitrary Control of the Initial Interface Shape

In this chapter, using magnetic levitation technique, we are able to generate an initial metastable interface between two immiscible fluids with a heavy fluid sitting on the top of a light fluid. When the magnetic field is turned off, both fluids are subject to an acceleration (gravity), allowing the Rayleigh-Taylor instability to occur. By affixing appropriately shaped magnetically-permeable wires to the outside of the cell, we impose arbitrarily-chosen and well-controlled initial perturbations on the interface. This technique is used to examine both the linear and nonlinear growth regimes for which the perturbation amplitudes, growth rates, nonlinear growth coefficients, and terminal velocities are obtained.
5.1 Introduction

In chapter 1, I described three stages of the RT instability. In the nonlinear regime, the growth rate declines due to nonlinearities [1–4]. In the final stage, the fluid interpenetration becomes turbulent, and memory of the initial perturbations fades or is lost [4–7]. It is the last two stages that receive the overwhelming bulk of attention, as there are significant inconsistencies among theory, simulation, and experiment [8]. This is due in large part to poorly defined initial conditions in experiments, particularly the presence of uncontrolled long-wavelength perturbations. To date experiments have been based mainly on physical motion of the cell [2, 5, 8–13]. These protocols suffer from jitter and not-well-controlled initial conditions, and tend to be poorly suited for establishing arbitrary initial interface perturbations that involve a specific single mode or spectrum of modes [11, 14]. In this chapter we demonstrate how an extension of our magnetic levitation technique [15, 16] enables us to fine-tune the initial conditions. We accomplish this by affixing precisely-shaped magnetically permeable materials to the outside of the fluid cell, which perturb the magnetic force and thereby the shape of the interface. On turning off the magnetic field, the total magnetic force disappears, leading to a purely gravity-driven RT instability with precisely controlled initial conditions. As a first application of this technique, we measure the amplitude of the initial interface perturbation and its growth due to an applied single mode perturbation as a function of the amplitude of a pair of sinusoidally-shaped magnetically-permeable wires.
Figure 5.1: Refractive index and density of the aqueous mixtures. Note that the refractive index $n$ was measured by using a Nd:YaG laser beam at 532 nm.

### 5.2 Working Fluids and Experimental Setup

In our experiments fluid 1 is a moderately strong paramagnetic mixture of water, 58.6 wt.-% $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$, approximately 1 wt.-% surfactant octa(ethylene glycol) dodecyl ether ("C$_{12}$E$_8$") to reduce surface tension, and a small amount of rhodamine 6G dye; fluid 2 is weakly diamagnetic hexadecane, which is immiscible with fluid 1 and of nearly the same refractive index. Fig. 5.1 shows the relationship among refractive index, density and concentration of $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$ aqueous mixture. The interfacial tension $\gamma = (2.6 \pm 0.3)$ erg cm$^{-2}$, as determined by the pendant drop technique (described in details in chapter 4), and wetting properties are such that
Figure 5.2: Cartoon of experimental setup. Note that no wires are attached to the outside of the cell, and that the x-axis view is expanded for clarity. The arrow indicating the direction of $\nabla (H^2)$ applies only in the absence of the wires.

the meniscus is virtually absent. When placed in a cell, the lighter hexadecane ($\rho_2 = 0.773 \text{ g cm}^{-3}$) ordinarily sits atop the heavier paramagnetic mixture (density $\rho_1 = 1.394 \text{ g cm}^{-3}$), where the Atwood number $A \equiv (\rho_1 - \rho_2) / (\rho_1 + \rho_2) = 0.29$. However, in the presence of a vertical magnetic field gradient $\nabla (H^2)$ [$\sim 2.0 \times 10^7 \text{ G}^2\text{cm}^{-1}$ at the interface], the denser fluid 1 resides above the less dense fluid 2 when $|\frac{1}{2} \chi_1 \nabla (H^2)| > |(\rho_1 - \rho_2) g|$; see Fig. 5.2. Here $\chi_1$ is the (positive) magnetic susceptibility of fluid 1 and the susceptibility of fluid 2 is negligible.

In order to control the initial perturbation of the RT instability, appropriately shaped paramagnetically-permeable wires were affixed to the outside of the cell, as shown in a schematic of the experimental setup in Fig. 5.3.
5.3 Data Acquisition and Results

On switching off the magnetic force, the layering becomes unstable in the presence of small, random perturbations — the large, controlled interfacial perturbations have not yet been applied — and the denser fluid 1 falls to the bottom of the container under uniform gravity. Fig. 5.5a (where “a” refers to the column) shows how the instability develops as a function of time for these fluids, with the initial observable growth being that of the fastest growing mode. In this experiment the cell is constructed of 0.2 cm thick glass; is 15 cm in height, 7 cm in width (span-wise, along the y-axis), and $d = 0.3$ cm thick (i.e., the cell gap along the x-axis); and has no observable meniscus (Fig. 5.5a-i). Note that the meniscus was checked by filling two fluids into a glass cuvette and observing the curvature of the interface between
two fluids. Here the interface is pretty flat.

Because the characteristic time for viscous diffusion across the cell [16] $t_v \sim d^2 \rho/\pi \eta \sim 0.5 \text{s}$, where $\eta \sim 5 \text{cP}$ is the mean viscosity, the system can be considered to be in the purely 2D RT regime for time $t \lesssim t_v$.

Using the technique of planar laser induced fluorescence (“PLIF” [17]), the cell is illuminated from above with a $\sim 0.1 \text{cm}$ thick “sheet” of light, created by passing a Nd:YaG laser beam at 532 nm through a cylindrical lens. The light sheet passes through the midplane of the cell, causing the dye in fluid 1 to fluoresce. Videos of the lower portion of the cell are collected at 60 frames per second using a CCD camera. (Simultaneous imaging of both upper and lower portions of the cell will become possible in the future with the addition of a second synchronized camera).

For the video acquisition, I make two remarks:

- Fluid 2 has nearly the same refractive index as fluid 1 to within 0.0003, and this refractive index match is very important because otherwise the interface between two fluids would be distorted.

- Also, in order to obtain a good quality video, a bandpass filter allowing the fluorescent light to pass was used to get rid of the green light of wavelength 532 nm, which is placed in front of the CCD camera.

A light-emitting diode (LED) is connected to the circuit through which a current is provided for the magnet. At the beginning, both the magnet and LED are on. On switching off the magnet, the RT instability occurs. The LED is used to determine
$t = 0$. When the LED intensity fast begins to decrease, this moment is defined as $t = 0$ in this experiment.

The instability passes first through the linear regime (Fig. 5.5a-ii) with a wavelength $\lambda^* = (0.69 \pm 0.05)$ cm for the fastest growing mode (denoted by an asterisk *), consistent with the prediction of $\lambda^* = 0.71$ cm from linear stability theory (LST) [18]. (We note that the measured $\lambda^*$ values also are nearly the same for cells of other thicknesses, viz., $d = 0.2$ and $d = 0.4$ cm). Note that the wavelength $\lambda^*$ was determined by averaging over the distances between two adjacent spikes in linear regime, as shown in Fig. 5.5a-ii.

Because of the small wavelength, video frames at early times have an insufficient number of pixels to resolve properly the initial growth rate $\sigma^*$, which LST predicts to be $33.5 \text{ s}^{-1}$. Using several well-resolved images at later times for which $h_k/\lambda^* \gtrsim 0.15$, where $h_k$ is the perturbation amplitude, we experimentally find $\sigma^* = (24 \pm 0.5) \text{ s}^{-1}$. As shown in Fig. 5.4, the initial growth rate $\sigma^*$ was determined by fitting to a linear function the logarithm of the amplitude $h_k$ as a function of time since $h_k = h_{k0}e^{\sigma^* t}$ in the linear regime. As expected, this is smaller than the LST prediction because the instability is already transitioning from the linear to the nonlinear regime, as seen in Fig. 5.5a-iii through Fig. 5.5a-vi.
5.4 Initial Perturbation Control and Data Analysis

Aside from physically agitating the cell to create standing waves [19] — a technique that severely limits the range of initial conditions and creates unwanted jitter — there is no viable extant method for establishing an arbitrary and controlled initial interface shape. Here we introduce a technique in which the initial fluid interface shape is manipulated by perturbing the magnetic field’s spatial profile. To accomplish this we use several pairs of T-304 cold-worked stainless steel wires (diameter = 0.088 cm) bent into a sinusoidal shape with a period of 2.25 cm and amplitudes $A_w = 0.10$ (Fig. 5.5b), 0.15, 0.20 (Fig. 5.5c), 0.25, and 0.30 cm (Fig. 5.5d). The wire’s relative magnetic permeability $\mu_r = (2.6 \pm 0.3)$, as measured by balancing the upward magnetic force against downward gravity (for details, see Appendix B).
Figure 5.5: Images of spike growth vs. time in sec., where $t = 0$ is the time at which the magnet current first begins to decrease. Column a) no wire; b-d) wires of amplitude $A_w = 0.1, 0.2, \text{ and } 0.3 \text{ cm}$, respectively.
Each pair of wires, painted black to reduce spurious light, is affixed to the outside of the cell’s two faces, 0.39 cm from the cell’s illuminated midplane (due to the thickness of the glass); see Fig. 5.3. The front wires can be seen as a silhouette in each image in Fig. 5.5b, c, and d; the rear wires are obscured and not easily visible. We emphasize that all aspects of the experiments shown in Fig. 5.5, including both the fluids and the cell, are identical, except for the addition of the wires in Fig. 5.5b, c, and d. [Note that the wires are placed so that the fluid interface is at the wires’ midpoint along z. The apparent vertical displacement in Fig. 5.5 is an illusion due to parallax]. On application of an external field the fluid interface is perturbed very slightly by the span-wise inhomogeneous magnetic force induced by the wires (Figs. 5.5b-i, 5.5c-i, and 5.5d-i), where the period of the interface instability \( \lambda = 1.12 \text{ cm} \) is exactly one-half that of the wires. Although too small to image, the amplitude \( h_{k0} \) of the initial perturbation at wavevector \( k \) can be estimated.

The amplitude \( h_k \) for the early growth can be derived from the equation [5,18,20]

\[
\frac{d^2 h_k}{dt^2} - \sigma^2 h_k = 0,
\]

where \( \sigma \) is approximately proportional to \( A^{1/2} \) [18]. However, because the magnetic force decays exponentially with a time constant \( \tau_m \sim 0.065 \text{ s} \), the effective Atwood number \( \sim A (1 - e^{-t/\tau_m}) \). Thus, we have

\[
\frac{d^2 h_k}{dt^2} - \sigma^2 \left(1 - e^{-t/\tau_m}\right) h_k = 0.
\]
Table 5.1: Interface perturbation amplitude $h_{k0}$, terminal velocity $(\frac{dh_k}{dt})_t$, and growth coefficient $\alpha_s$ for different wire amplitudes $A_w$.

<table>
<thead>
<tr>
<th>$A_w$ (cm)</th>
<th>No wire</th>
<th>0.1</th>
<th>0.15</th>
<th>0.2</th>
<th>0.25</th>
<th>0.3</th>
</tr>
</thead>
<tbody>
<tr>
<td>$10^6 \times h_{k0}$ (cm)</td>
<td>0.043 ± 0.008</td>
<td>0.4 ± 0.1</td>
<td>0.9 ± 0.2</td>
<td>1.6 ± 0.4</td>
<td>3.9 ± 0.9</td>
<td>6 ± 1</td>
</tr>
<tr>
<td>$(\frac{dh_k}{dt})_t$ (cm s$^{-1}$)</td>
<td>8 ± 0.5</td>
<td>11 ± 1</td>
<td>11 ± 1</td>
<td>10 ± 2</td>
<td>10 ± 1</td>
<td>11 ± 1</td>
</tr>
<tr>
<td>$\alpha_s$</td>
<td>0.065 ± 0.01</td>
<td>0.06 ± 0.02</td>
<td>0.07 ± 0.01</td>
<td>0.06 ± 0.02</td>
<td>0.06 ± 0.01</td>
<td>0.07 ± 0.02</td>
</tr>
</tbody>
</table>

The solution to the above Eq. (5.2) is

$$h_k = h_{k0} J_{-\beta} \left( \beta e^{-t/2\tau_m} \right) / J_{-\beta}(\beta),$$

(5.3)

where $J_{-\beta}$ is a Bessel function of the first kind of order $-\beta$ and $\beta = 2\sigma\tau_m$, Eq. (5.3) is equal to $h_{k0} \exp(\sigma t)$ in the limit $\tau_m \to 0$. Fitting $h_k$ to the two earliest resolvable interface amplitudes $h_k(t)$ (such that $h_k \sim 0.5$) for each wire amplitude $A_w$, along with the LST values $\sigma = 33.0$ s$^{-1}$ (cells with wires, $k = 5.6$ cm$^{-1}$), we determine $h_{k0}$ (Table 5.1) and the dimensionless quantity $kh_{k0}$ (inset in Fig. 5.6). As an example, Fig. 5.7 shows two earliest resolvable amplitudes ($h_k$) were fitted to obtain the initial amplitude $h_{k0}$ for wire amplitude $A_w = 0.25$ cm.

These values are considerably larger than those that occur without the perturbing wires (Table 5.1), where we use the LST value $\sigma^* = 33.5$ s$^{-1}$ (cell without wires, $k^* = 9.1$ cm$^{-1}$). Additionally, the data indicate that $h_{k0}$ increases smoothly with $A_w$. When the field is switched off, Fig. 5.5b-ii through vi, Fig. 5.5c-ii through vi, and Fig. 5.5d-ii through vi show the evolution of the perturbations. (In fact, although not shown here, data were collected for a total time $t \sim 30/\sigma^*$ s before the spikes reached the bottom of the cell. This is important for the study of the late time behavior because the instability evolves into the late time regime at about 0.35 s and only after this time it is possible to observe some interesting behavior such as
Figure 5.6: Main figure: $\frac{|\vec{F}_{\text{wire}} - \vec{F}^0|}{|\vec{F}^0|}$ in the midplane of the cell for $A_w = 0.25$ cm. Wide gray curve is projection of wire into the midplane, dotted line is fluid interface at $t = 0$, and solid curve is actual fluid interface measured at $t = 0.265$ s. The force, which affects fluid 1 due to its large $\chi_1$, is largest near the wire’s crossing points, and results in a collection of fluid 1 at the crossing points. Inset: Dimensionless interface perturbation amplitude $kh_{k_0}$ vs. wire amplitude $A_w$. 
Figure 5.7: Two earliest resolvable interface amplitudes fitted with Eq. (5.3) to obtain $h_{k0}$ for wire amplitude $A_w = 0.25$ cm.
Figure 5.8: Growth data vs. time for cell with wire of amplitude $A_w = 0.3$ cm. a) the position of the front, b) the velocity of the front, and c) the instantaneous growth coefficient $\alpha_s$.

Several features are apparent. First, as noted above, the wavelength of the interface instability is half that of the wire, with spikes appearing where the interface crosses the wires; this will be discussed below. Second, the spikes arising from initially larger amplitude perturbations of the fluid interface in Fig. 5.5 grow to correspondingly longer lengths (i.e., larger $h_k$) before the instability evolves into the late time regime at about $t \sim 0.6$ s. This behavior occurs because of the larger initial amplitudes $h_{k0}$, which can grow significantly before interaction with other modes becomes important.

Fig. 5.8a shows the advance $h_k$ vs. time of the spike front for the $A_w = 0.3$ cm
Figure 5.9: Growth data vs. time for cell without wires. a) the position of the front, b) the velocity of the front, and c) the instantaneous growth coefficient $\alpha_s$.

amplitude wire (Fig. 5.5d), as defined by the maximum extent to which least 5% of the denser fluid has fallen. Fig. 5.8b shows its velocity $\frac{dh_k}{dt}$, and Fig. 5.8c shows the instantaneous growth coefficient $\alpha_s$, defined as $[23, 24]$ 

$$(dh_k/dt)^2 = 4A\alpha gh_k \text{ or } \alpha = (dh_k/dt)^2 / 4Agh_k,$$  \hspace{1cm} (5.4)

where $\alpha$ can be $\alpha_s$ or $\alpha_b$, and $g$ is the gravitational acceleration. As a comparison, Fig. 5.9a shows the advance $h_k$ vs. time of the spike front for the cell without a wire (Fig. 5.5a); Fig. 5.9b shows its velocity $\frac{dh_k}{dt}$, and Fig. 5.9c shows the instantaneous growth coefficient $\alpha_s$.

Eq. (5.4) used to determine the same form for $\alpha$ was recently obtained by Ristorcelli and Clark [25] and Cook et al. [23, 26], although they used different
approaches. Ristorcelli and Clark used a similarity assumption that the solutions to the momentum equations can be expressed to be the product of a temporal scaling function and a spatial similarity function, whereas Cook et al. assumed the mass flux and energy balance. The solution to Eq. (5.4) is [26]

$$h_k(t) = \alpha Ag t^2 + 2(\alpha Ag h_k)^{1/2} t + h_{k0},$$

(5.5)

Another technique introduced by Dimonte et al. [27, 28] is to measure $\alpha$ by plotting $h_k$ versus $Ag t^2$ and fitting a line to the most visually pleasing portion of the data. Compared to $\frac{h_k}{Ag t^2}$ from Eq.(5.5), the difference between these two techniques can be very big, as seen in Fig. 5.10.

Figure 5.10: Comparison of measurement techniques for the growth rate coefficient $\alpha$. Courtesy to Cabot et al. [26] for this figure where $\tau$ is a natural timescale.
\( \alpha \) has been the subject of intense interest \([8,29]\), as there are significant discrepancies between experiments, which in the past have been plagued by not-less-controlled initial conditions, and calculations, which depend critically upon the initial perturbation spectrum. In a potential flow model involving mode competition, Birkoff \([30]\) obtained a value of \( \alpha_b = 0.06 \). According to bubble merger models \([4,21,31,32]\), \( \alpha_b \) values are possibly independent of the initial conditions but depend on merger rate. However, Dimonte \([27]\) showed that \( \alpha_b \) can change by a factor of 2-3 depending on initial conditions. By introducing an artificial interface with a perturbation spectrum centered on the most unstable mode, continuum simulations generally yield values for \( \alpha_b \) and \( \alpha_s \) that are only about half of those observed in experiments, as shown in Table 5.2. Kadau et al. \([33]\) attributed these low values to the absence of thermal fluctuations for the initial interface. In the presence of the fluctuations, their atomistic simulations are qualitatively in good agreement with the experimental results \( \alpha_b = 0.063 \) \([5,34]\). Adopting a broadband perturbation spectrum on the initial perturbations, Ramaprabhu et al. \([29]\) also obtained large values of \( \alpha_b \).

We note from Fig. 5.8c that \( \alpha_s \sim (0.07 \pm 0.02) \) during the self-similar growth period \( t = 0.3 \) to \( t = 0.45 \) s, after which it decays as the velocity has reached a terminal value \( \left( \frac{dh}{dt} \right)_t \sim 11 \) cm s\(^{-1} \) (Fig. 5.5d-iii). Although a terminal velocity is expected to occur for small \( A \) when only a single mode is present \([12,38]\) and is observed in all of our measurements, we believe that the origin of the terminal velocity lies elsewhere. That the crossover from self-similar growth to terminal velocity occurs in all cases in the neighborhood of \( t \sim 0.5 \) s, which corresponds
Table 5.2: Growth coefficients for bubbles ($\alpha_b$) and spikes ($\alpha_s$).

<table>
<thead>
<tr>
<th>$A$</th>
<th>Atomistic [33]</th>
<th>Experiment [35]</th>
<th>Continuum</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha_b$</td>
<td>0.061 (3D, DSMC)</td>
<td>0.051 (3D [35])</td>
<td>0.031 (3D [26])</td>
</tr>
<tr>
<td></td>
<td>0.059 (2D, DSMC)</td>
<td>0.063 (3D [5])</td>
<td>0.028 (2D [36])</td>
</tr>
<tr>
<td></td>
<td>0.051 (2D, MD)</td>
<td>0.063 (3D [37])</td>
<td></td>
</tr>
<tr>
<td>$\alpha_s$</td>
<td>0.29</td>
<td>0.070 (2D [this work])</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.43</td>
<td>0.067 (3D [35])</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.50</td>
<td>0.070 (3D [35])</td>
<td>0.031 (3D [37])</td>
</tr>
<tr>
<td></td>
<td>0.50</td>
<td></td>
<td>0.034 (2D [36])</td>
</tr>
<tr>
<td></td>
<td>0.67</td>
<td>0.074 (2D, DSMC)</td>
<td>0.084 (3D [35])</td>
</tr>
<tr>
<td></td>
<td>0.75</td>
<td>0.086 (2D, MD)</td>
<td>0.097 (3D [35])</td>
</tr>
<tr>
<td></td>
<td>0.90</td>
<td>0.103 (2D, DSMC)</td>
<td>0.133 (3D [35])</td>
</tr>
</tbody>
</table>

to the diffusion time $t_v$ for the surface vorticity layer, suggests that the terminal velocity is due in part to the instability’s transitioning from the 2D RT to the Hele-Shaw regime [16]. We emphasize that the initial linear growth regime and most of the subsequent self-similar growth regime, both taking place before vorticity has reached the mid-plane of the cell, should thus be very little influenced by friction on the glass walls. It also has been suggested that fluctuations result in a coarsening of the spikes, resulting in droplet breakoff, after which droplets are subject to Stokes-like frictional flow with a terminal velocity [33].

5.5 Control of the Wire Shape by Calculating the Magnetic Force Profile

I now describe the calculation by colleague Dr. Tim Atherton for the static force on the fluids in the presence of the wires. In the absence of current a scalar magnetic
potential $\varphi$ may be introduced into Laplace’s equation

$$\nabla \cdot (\mu_r \nabla \varphi) = 0,$$

(5.6)

where $\vec{H} = -\nabla \varphi$. A commercial finite-element package, FlexPDE, is used to solve this equation on a computational domain of physical dimensions $3.0 \times 2.25 \times 4.0$ cm, with periodic boundary conditions. In the model the wires are placed at the same positions relative to the fluid interface as in the experiment, and with sinusoidal shape having $A_w = 0.25$ cm and period $2.25$ cm. Wires of relative permeability $\mu_r = 2.6$ and square cross-section are used in the calculation, such that the wires have a cross-sectional area equal to those used in the experiment. Dirichlet conditions are applied at the pole pieces. The total magnetic force $\vec{F}_{\text{wire}}^{\text{wire}} \left[ \propto (\vec{H} \cdot \nabla) \vec{H} \right]$ with the wires present minus the force $\vec{F}_0$ in the absence of the wires, scaled by $\vec{F}_0$, at the cell’s mid-plane is plotted in Fig. 5.6; also shown is the experimental interface at $t = 0.265$ s. It is clear that the wires produce a force on paramagnetic fluid 1 that is directed toward the wire’s crossing points with the interface. Thus the overall energy, including magnetic, gravitational, and surface tension, can be reduced if the paramagnetic (upper) fluid collects near the wire / fluid interface crossings, lowering the interface near the crossings and raising it near the wires’ extrema. For small wire amplitude $A_w$, the shape of the interface is very nearly a single sinusoidal mode, as seen in Fig. 5.6. With increasing $A_w$, however, contributions from higher harmonics of $k$ begin to emerge, suggesting that $h_{k0}$ can be increased more efficaciously by increasing $\mu_r$ or the wire diameter.
The interface profile can be determined from the magnetic force. For instance, Fig. 5.6 shows the distribution of the magnetic force on the interface for $A_w = 0.25$ cm. The force is approximately perpendicular to the wire in the $y$-$z$ plane. Because there is a component of the force pointing to the wire’s crossing points with respect to the interface, at these points a concave shape occurs, as also shown in Fig. 5.6.

5.6 Summary

To summarize, we have studied the growth of an imposed single mode perturbation. One also can create an arbitrary spectrum of initial perturbation modes. This requires the solution of the inverse problem in which the appropriate magnet current, wire shape, and permeability need to be determined to achieve the desired initial perturbation profile. Besides demonstrating a powerful technique for RT experiments with arbitrary initial conditions, this work has yielded a precise measurement of the growth coefficient $\alpha_s$, which is independent of wire amplitude for a precise set of experimental condition: a moderately large Atwood number ($A = 0.29$) and a jitter-free single-mode initial perturbation, two elements that would have been difficult to achieve by other means. Clearly, the ability to manipulate the initial interface shape facilitates many heretofore inaccessible investigations involving controlled initial conditions.

References


[33] K. Kadau, C. Rosenblatt, J.L. Barber, T.C. Germann, Z. Huang, P. Carlès, 


Pylaev. In Proceedings of the third International Workshop on the Physics of 
Compressible Turbulent Mixing, ed Dautray R (Commissariat Energie Atom-

Appendix A

Digitization of the Interface between Two Fluids in an Image

In this appendix, a method used to detect an edge in an image is presented. The goal of the edge detection is to mark the points in an image where the luminous intensity varies sharply.

A.1 Algorithm in Canny Edge Detection

Standard edge detectors, such as the Sobeland Prewitt detectors, are finite-difference-based first-derivative operators, which only pick up high-frequency responses at image edges. Recently, Canny [1] formulated edge detection as an optimization problem, and the Canny detector can be effectively approximated by the first derivative of the Gaussian function. His idea has been developed and improved by many authors [2–7].
A.1.0.1 Image Smoothing

First of all, an image is smoothed by a 2-D Gaussian filter given by

\[ G(x, y) = \frac{1}{2\pi\sigma^2} e^{-\frac{x^2 + y^2}{2\sigma^2}}, \]  

where \( \sigma \) is a user-defined parameter.

Using a gaussian filter, the convolution \( g(x, y) \) is obtained from the original image \( f(x, y) \) as follow

\[ g(x, y) = G(x, y) \otimes f(x, y). \]  

where \( \otimes \) is the convolution operator defined as

\[ G(x, y) \otimes f(x, y) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dx'dy'G(x', y')f(x - x', y - y'). \]  

Normally, two dimensional convolution \( g(x, y) \) with large Gaussians takes a long time, in practice the 2-D convolution can be performed by first convolving with a 1-D Gaussian in the \( x \) direction, and then convolving with another 1-D Gaussian in the \( y \) direction. This produces two (rather than one) values at each pixel. This first step seems to be necessary because the noise in an image will be reduced after a Gaussian smoothing.

A.1.0.2 Gradient Operator

Assume that two 1-D convolutions have been obtained during a Gaussian smoothing. Then the values of the \( x \)-smoothed image array are convolved with a first
derivative of a one dimensional Gaussian of identical aligned with $y$. Similarly, values in the $y$-smoothed image array are convolved with a first derivative of a one dimensional Gaussian of identical aligned with $x$.

The smoothed image $g(x, y)$ is differentiated along $x$ and $y$ direction, respectively. Then we have the $x$- and $y$-component gradients

$$g_x(x, y) = \frac{\partial g(x, y)}{\partial x} = \frac{\partial G(x, y)}{\partial x} \otimes f(x, y), \quad (A.4)$$

$$g_y(x, y) = \frac{\partial g(x, y)}{\partial y} = \frac{\partial G(x, y)}{\partial y} \otimes f(x, y). \quad (A.5)$$

The magnitude of the gradient of $g(x, y)$ and the angle of its slope can be calculated as follows

$$M(x, y) = \sqrt{g_x^2(x, y) + g_y^2(x, y)}, \quad (A.6)$$

$$\theta(x, y) = \tan^{-1} \left[ \frac{g_y(x, y)}{g_x(x, y)} \right]. \quad (A.7)$$

A.1.0.3 Non-maximum Suppression

The purpose of Non-maximum suppression is to thin the edge ridges. To perform this, each of non-zero $M(x, y)$ is checked and compared to its neighbors along the $\theta(x, y)$ direction. If it is bigger than the neighbors, then $M(x, y)$ is kept unchanged; otherwise, it is set to zero.
A.1.0.4 Threshold

In most cases, a single threshold limit $T$ is used to select the points. When a value of a point (let’s say, intensity) is above the threshold, it will be accepted. However, the single threshold limit will produce a streaking (a broken line) when the edge values fluctuate above and below the threshold. Thus, to avoid the streaking, a method called “hysteresis” is used to threshold $M(x, y)$. In this method, two threshold limits (upper and low limits) are used to select the points. Considering a line segment, if a value of a point lies above the upper threshold limit, the point is immediately accepted; otherwise, it is immediately rejected. Points which lie between the two limits are also accepted if they are connected to pixels which exhibit strong response. In other words, these points have much higher intensity than does the noise. In such a way, the likelihood of streaking is reduced drastically. It is recommended that the ratio of the upper to the low limit be in the range two (or three) to one, based on predicted signal-to-noise ratios [1].

After performing threshold for $M(x, y)$ by two different threshold limits $T_1$ and $T_2$ ($T_2 > T_1$), two new images $M_{T_1}(x, y)$ and $M_{T_2}(x, y)$ are obtained as follows:

$$M_{T_1}(x, y) = \begin{cases} M(x, y) & \text{if } M(x, y) \geq T_1; \\ 0 & \text{otherwise.} \end{cases} \quad (A.8)$$

$$M_{T_2}(x, y) = \begin{cases} M(x, y) & \text{if } M(x, y) \geq T_2; \\ 0 & \text{otherwise.} \end{cases} \quad (A.9)$$

The threshold $T_1$ is so chosen that the edge points are kept while most of the noise is suppressed. Note that the image $M_{T_2}(x, y)$ has less noise, fewer false edge points.
and larger gaps between the edge segments than \( M_{T_1}(x, y) \). The final stage is to link the edge segments in \( M_{T_2}(x, y) \) to form a continuous contour of edges. This can be done by tracing each segment in \( M_{T_2}(x, y) \) to its end and then searching its neighbors in \( M_{T_1}(x, y) \) to find any edge segment in \( M_{T_1}(x, y) \) to bridge the gap in \( M_{T_2}(x, y) \) until reaching another edge segment in \( M_{T_2}(x, y) \).

### A.1.1 Results

Based upon Canny’s criteria, we are able to analyze all frames in a video. Here I should point out that only one threshold limit \( T \) was used in the edge detection. If we use a lower threshold limit and an upper one, the quality of the edge detection would be improved. Nevertheless, at this moment we are satisfied with the results by using one threshold limit.

![Figure A.1: A frame from a video.](image1.png)

![Figure A.2: An edge profile obtained after performing an edge detection.](image2.png)

As an example, an original image extracted from a video is shown in Fig. A.1. Fig. A.2 shows an image after the above algorithm is performed. Note that the points near both horizontal ends of the image are noisy due to the lack of pixels.
close to the ends. When the digitized edge profile is plotted into the original frame, a new image is obtained, as shown in Fig. A.3.

**A.1.2 Summary**

To summarize, the algorithm of Canny edge detection is presented. Based on Canny’s criteria, an edge detection has been used to digitize the interface between two fluids in our experiment. The natural work in the future will be to use two threshold limits to improve the results of the digitized profile.

**References**


Appendix B

The Measurement of the Relative Magnetic Permeability of a Material

In this appendix, the method used to measure the relative magnetic permeability is presented. First of all, by solving the boundary value problem for an infinite cylinder of magnetic permeable material in a uniform external magnetic field \( H_0 \) perpendicular to the cylinder’s axis, the magnetic force due to the magnetic dipole moment of the wire is obtained. Later on, the downward gravitational force on the wire is canceled by the upward magnetic force, and thus the magnetic permeability of the wire is obtained.

B.1 The Solution to the Boundary Value Problem

For an infinite cylinder with radius of \( a \) in a uniform magnetic induction \( B_0 \) shown in figure B.1, by using cylindrical coordinate system, the potential satisfying
Figure B.1: an infinite cylinder in a uniform magnetic field.

Laplace equation can be expressed as follow,

$$\nabla^2 \varphi = \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \varphi}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2 \varphi}{\partial \phi^2} = 0 \tag{B.1}$$

The general solution to the equation B.1 is as follow:

$$\varphi = \sum_{n=0}^{\infty} \left( C_n r^n + D_n r^{-n} \cos(n\phi) \right) \tag{B.2}$$

In order to obtain the coefficients $C_n$ and $D_n$, one has to use the following boundary conditions:

1. as $r \to 0$, $\varphi$ is finite, then one has

$$\varphi_1 = \sum_{n=0}^{\infty} C_n r^n \cos(n\phi), \quad r \leq a \tag{B.3}$$
2. as \( r \to \infty, \varphi \to -\frac{B_0}{\mu_0} r \cos(\phi) \), hence

\[
\varphi_2 = \sum_{n=0}^{\infty} \frac{D_n}{r^n} \cos(n\phi), \quad r \geq a
\]  

(B.4)

3. when \( r = a, B_{1n} = B_{2n} \), that is to say,

\[
-\mu \left( \frac{\partial \varphi_1}{\partial r} \right)_{r=a} = -\mu_0 \left( \frac{\partial \varphi_2}{\partial r} \right)_{r=a}
\]  

(B.5)

One finally can obtain the coefficients as follow,

\[
C_n = 0, \quad D_n = 0, \quad \text{for } n \neq 1
\]  

(B.6)

\[
C_1 = -\frac{2}{\mu + \mu_0} B_0, \quad D_1 = \frac{\mu - \mu_0}{(\mu + \mu_0)\mu_0} B_0
\]  

(B.7)

Therefore, the potential is obtained as follow,

\[
\varphi_1 = -\frac{2}{\mu + \mu_0} B_0 r \cos \phi, \quad r \leq a
\]  

(B.8)

\[
\varphi_2 = \frac{\mu - \mu_0}{\mu + \mu_0} a^2 B_0 \frac{r}{r} \cos \phi - \frac{B_0}{\mu_0} r \cos \phi
\]  

(B.9)

\[
= \left[ \frac{\mu - \mu_0}{\mu + \mu_0} \left( \frac{a}{r} \right)^2 - 1 \right] \frac{B_0}{\mu_0} r \cos \phi, \quad r \geq a
\]  

(B.10)

Thus, the magnetic induction can be obtained as follow

\[
B_1 = \mu H_0 = \mu \nabla \varphi_1 = \frac{2\mu}{\mu + \mu_0} B_0, \quad \text{for } r \leq a
\]  

(B.11)

\[
B_2 = B_0 + \frac{\mu - \mu_0}{\mu + \mu_0} B_0 \left( \frac{a}{r} \right)^2 (\cos \phi \hat{r} + \sin \phi \hat{\phi}), \quad r \geq a
\]  

(B.12)
B.2 Magnetic Force on a Wire

Now let’s consider a finite cylindrical wire with a radius of \(a\) in the magnetic field \(H_0\) and use cgs unit. Inside the cylinder the magnetic field \(H\) is uniform and is equal to \(\frac{2}{1+\mu_r} H_0\). Thus, one has \(B = H_0 \left(\frac{2\mu_r}{1+\mu_r}\right)\).

The magnetization inside the cylinder is obtained as follow

\[
M = \frac{B - H}{4\pi} = \frac{H_0 \mu_r - 1}{2\pi \mu_r + 1}
\]  

(B.13)

Total magnetic dipole moment is

\[
m = M \times \text{volume} = \frac{H_0 \mu_r - 1}{2\pi \mu_r + 1} L \pi a^2
\]  

(B.14)

where \(L\) is the length of the wire.

The magnetic force exerted on the wire is

\[
F = \frac{1}{2} \nabla (m \cdot B_0) = \frac{1}{2} m \nabla B_0
\]  

(B.15)

\[
= \frac{1}{4\pi \mu_r + 1} L \pi a^2 H_0 \nabla H_0
\]  

(B.16)

where \(B_0\) is the magnetic induction without the wire in place. Note that the factor \(\frac{1}{2}\) is due to the energy density of a localized distribution of permanent magnetization in an external field rather than a permanent magnetization [1].

B.3 Measurement of Relative Permeability

Now turn to the experimental measurement for the relative permeability. The
Figure B.2: A schematic of the experimental setup for relative permeability measurement.
experimental setup is shown in figure B.2. We cut a piece of wire \( L = 2.199 \text{ cm} \) in length, and the radius \( a = 0.044 \text{ cm} \) [note that the ratio \( L/a \) is more than 40, which is why we felt comfortable doing the original calculation for the infinite cylinder case]. When placed in a field gradient, the wire experiences a upward force given by equation B.16. Note that in fact the field cannot be uniform, otherwise we cannot do the experiment. But over the size of the wire the field does not change much. In order to measure the relative permeability \( \mu_r \) at values of magnetic field at which we performed the Rayleigh-Taylor instability experiment, we wrapped a nonmagnetic thread (normally used for sewing) around the wire segment and placed the wire segment on a V-shaped platform in the magnet at the point where the upward force is largest and spatially uniform. We slipped the thread through a hole in the platform and let it hang well below the magnet. At the end of the thread we hung a vial. We turned up the current to 20 A, near which we did the Rayleigh-Taylor instability experiments, and the wire/thread/vial levitated. We then added enough water to the vial so that the wire/thread/vial/water would no longer levitate, and then weighed the wire/thread/vial/water to determine the gravitational force that was canceled by the magnetic force corresponding to \( \nabla B_0 = (2.32 \pm 0.01) \times 10^3 \text{ G cm}^{-1} \) at this current (\( I = 20 \text{ A} \)).

It turned out the relative permeability of the wire in our experiments is 2.6. Although this number is high for an ordinary stainless steel, which tends to be \( \mu_r \sim 1.005 \), we are working with “cold worked” stainless steel which can have values of \( \mu_r \) in the neighborhood between 1.5 and 2.3 when measured in fields of 200
Oersteds (Oe). At much higher fields (around 8000 Oe) at which the measurement was carried out and at which the Rayleigh-Taylor instability experiments have been performed, $\mu_r$ will be larger.

In a word, we showed a useful method for measurement of relative permeability using magnetic levitation technique.

**References**

Appendix C

Mapping of Images Distorted by Optical Parallax

In this chapter, we talk about the way of mapping the actual images distorted by optical parallax onto a nice rectilinear grid, which is how the experiment actually proceeds. Using Matlab, we converted the old \((x, y)\) coordinates from the original image to new \((x', y')\) rectilinear coordinates, and then for each new point \((x', y')\) with integer values, went back to the original image and determined the corresponding non-integer \((x, y)\) values. From this, we interpolated the intensity in the video at this non-integer \((x, y)\) point using the intensities at the integer \((x, y)\) points around it. It is this interpolated intensity that we assign to the new rectilinear \((x', y')\) coordinate. This procedure is accurate, but has the effect of very slightly washing out what might otherwise be a sharp border, that otherwise would have a large step change in intensity from one pixel to the adjacent pixel. Nevertheless, we suspect that it is the best possible procedure.
Figure C.1 showed the original image with optical parallax included. One has to find the symmetry axis along which one has $f(-x) = f(x)$. It can be done by extending the two lines that define the left and right edges and finding the x-coordinate of the intersection point. For example, the linear equations for the lines labeled 5 and 6 in figure C.2 are

$$y = 106.41x - 41475, \quad (C.1)$$

$$y = -90x + 29741 \quad (C.2)$$
and so the intersection point is $(363, -2929)$. Now one must obtain the horizontal distance between the line labeled 1 and the symmetry axis as a function of vertical position. To do this, many points $(x_{\text{pixel}}, y_{\text{pixel}})$ may be taken from the image along the line 1 and then fitted to a low-order polynomial function yielding length $L$ as a function of $y_{\text{pixel}}$. For line 1 in the figure C.2, one has $L(y) = -5.707 \times 10^{-5} y^2 + 0.10912y + 264.18$.

A trapezium (in bold) in the raw frame in figure C.3 is mapped onto a rectangle (in bold) in the processed frame as described above. It is therefore necessary to locate the $x$-coordinates of the left and right hand vertices at the top of the trapezium. Drawing a line between the intersection point $(363, -2929)$ and the point $(1, 494)$ located at the bottom left corner in figure C.2, one can locate the top left hand
Figure C.3: a trapezium in the raw frame mapped onto a rectangle in the processed frame.

corner of the trapezium with coordinates (52, 1); this point will be mapped to the coordinate (1, 1) in the processed frame. The top-right corner of the trapezium (615, 1) is found in a similar manner. Thus the size of the new map (in red) in figure C.3 is 494 × 564 (the number of pixels in y, by the number of pixels in x).

It is helpful to translate the image coordinates at this point so that the symmetry axis (the blue line in figure C.2) lies along $x'' = 0$. In these translated $(x'', y'')$ coordinates, the left boundary is $x''_{left} = 52 - 363$ and the right boundary $x''_{right} = 615 - 363$. The relation of the coordinates between the raw frame and the processed frame is $(x = 363 + x'/R)$, where $R = L(y' = 1)/L(y')$.

The intensity value of each pixel in the processed frame must now be found from the raw frame. For each pixel in the processed frame, the coordinates of the corresponding point in the raw frame are found. Since in general the $x$-coordinate of this point is non-integer, the intensity assigned to the pixel in the processed frame must be obtained by some interpolation. We choose a linear interpolation so that
the intensity of a pixel in the processed frame at coordinates \((x', y')\) is

\[
I_{\text{new}}(y', x', i) = I_{\text{old}}(y, x_{\text{int}}, i) \ast (x_{\text{int}} + 1 - x) + I_{\text{old}}(y, x_{\text{int}} + 1, i) \ast (x - x_{\text{int}}),
\]

where \(i\) represents one of the three orthogonal RGB color components. After the horizontal correction, one has the image without the horizontal distortion as shown in figure C.4.

\[\text{Figure C.4: the image without the horizontal distortion.}\]

\[\text{horizontal correction, one has the image without the horizontal distortion as shown in figure C.4.}\]

\section*{C.2 Vertical Correction}

Now one has to correct the vertical distortion. By measuring the vertical length of the grids as a function of vertical position, one obtains the difference between the real dimension and the vertical length in the image, listed in table C.1:
Table C.1: the difference between the real dimension and the vertical length in the image due to the distortion.

We know in real dimension the size of each grid is 1 cm corresponding to 53 pixels. Using the function

\[ y_{\text{image}} = y_{\text{real}} + ay_{\text{real}}^2 \]  

(C.4)

to fit the data in table C.1, one obtains the coefficient \( a \) (for instance, here \( a = 3.5189 \times 10^{-4} \)). Similarly, one then performs the intensity matching as the above but along the vertical direction.

\section*{C.3 The Image after Optical Parallax Correction}

Now check the new image after the full \((x, y)\) mapping, one notices that each grid is not a square, with 53 pixels along the vertical direction and 58 pixels along the horizontal direction. Use the ratio \( R = 53/58 \) to re-scale \( x \) coordinates and obtain a new map with the grid being a square, as shown in figure C.5.
Figure C.5: The new image without optical parallax.
Appendix D

Procedure on Performing the Rayleigh-Taylor Instability Experiment

In this appendix, I’ll present the procedure on the RT instability experiments. In order to obtain a good quality video, a clean cell is required.

D.1 Cell Cleaning

Assume that a cell has been made using pieces of glass. Since the side walls of the cell are sealed with ten minute black epoxy, it is necessary to pay attention to using solvents such as acetone to avoid the leak during the cleaning.

- Dip a home-made Q-tip into acetone and then use the Q-tip to wipe the inside surfaces of the cell.

- Fill the cell with ethanol and then use a home-made Q-tip to wipe the surfaces of the cell.
Repeat the above steps 3-4 times.

Use an air gun to dry the cell.

See if the cell is clean by checking whether there is any remanent on the inside surfaces of the cell.

Repeat the above steps if the cell is not clean.

D.2 Alignment of Two Pole Pieces

By using a bubble level, two pole pieces of the magnet are aligned such that the magnetic force produced by a current is vertically upward.

D.3 Setup

With the magnetic field on, the cell is placed between the two pole pieces of the magnets, as shown in Fig. D.1. Note that the offset in the function generator is about 0.4 volts. The reason to turn on the magnets is that the alignment of two pole pieces will be kept the same as before.

D.4 Filling with the Fluids

First, the light fluid (hexadecane) is filled with a home-made needle (a regular needle is replaced with a 15 cm long stainless steel tubing) into the lower half of the cell. Secondly, with the magnetic field on (the dial of the offset in the function generator is 1.2 volts, corresponding to the current 24.2 A in a circuit with a resistor
of 1.91 \( \Omega \)), the heavy fluid (an aqueous mixture of 58.6-wt.% \( \text{MnCl}_2 \cdot 4\text{H}_2\text{O} \), 1-wt.% \( \text{C}_{12}\text{E}_8 \) and a tiny amount of rhodamine 6G dye) is filled with a home-made needle on top of the light fluid, as shown in the upper part of Fig. D.1. Finally, a microscopy slide covered with a black tape (with a 1 mm wide opening slit) is used to eliminate the meniscus at the air interface such that the heavy fluid touches the cover slice without any air bubble left inside the cell. The slit is useful because only 1 mm wide laser beam is allowed to pass.

**D.5 Video Acquisition**

The current is reduced to 22.4 A corresponding to 1.02 volts—the dial of the offset in the function generator. After waiting for about 20 minutes to minimize the meniscus of the interface between the two fluids, one can begin to take a video.
Bibliography

http://www.lci.kent.edu/lc_history.html.


