Defects and Distortions of Layered Complex Fluids

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1 Overview of the Field

Layers abound in many important media: many kinds of biological membranes consist of lipid bilayers and form many structures important for cell biology, including organelles and cell membranes. Dense suspensions of elongated solid particles, such as viruses, bacteria and colloidal particles, may form layered configurations in small clusters or with long-range order. Smectic liquid crystals are a paradigmatic example of a fluid with internal layered order. Block copolymers, immiscible mesogens bonded together, form layered phases as just one of many possible configurations.

In each of these materials, the geometric and topological consequences of layering play a key role in the mechanics and solution space of configurations. Membranes form sheets, tubules, vesicles and helicoids as a consequence, for example. In smectic liquid crystals, where the layer spacing is approximately constant, the structures adopted are limited to a few families of surfaces and readily allow the phase to be identified under a microscope. External influences, such as boundary conditions and applied fields, may force deformations of the fluid that are incompatible with the layering, leading to geometric frustration and the spontaneous assembly of a wide variety of textures with characteristic defect structures.

Early interest in layered media, especially smectics, was driven by a broader interest in critical phenomena, i.e. the nature of phase transitions and how they are characterized. A notable example is the de Gennes description of the nematic-smectic A transition, involving a complex order parameter,

$$\psi = \delta\rho e^{iqn\cdot r}$$

where $\delta\rho$ is the amplitude of the density modulation, q is the wavenumber, n is the layer normal and r is position. The theory built from this yielded a Nobel-Prize-winning analogy to the normal metal-superconducting transition. An important consequence of this analogy is that vortex lattices observed in superconductors have a liquid crystal analogy, the twist-grain-boundary phase, which was subsequently discovered experimentally.

Common approaches to understanding the structure of layered media have therefore tended to focus on explaining the origin of layering and exploiting differential geometry methods to understand the wealth of structures that arise. In contrast, layered media have proved remarkably challenging to model computationally. Such methods could be of great benefit to structure prediction, particularly in scenarios where partial smectic order exists, such as during phase transitions, or to understand dynamical phenomena such as the shape evolution of films and bubbles.

The primary objective of this workshop was, therefore, to bring together computational physicists, applied mathematicians and experimentalists to identify: Geometries and experimental phenomena of interest that have become amenable to simulation; New theoretical questions about the structure and self-assembly of layered media that could be investigated computationally; New experiments that may now be possible due to emerging computational modelling approaches; Opportunities to formulate simulation methods for other layered media beyond smectics; New mathematical approaches that might further accelerate research on the broad category of layered fluids; and, How to exploit the interplay of geometry, topology and computation for improved algorithms.

2 Recent Developments and Open Problems

The conference was motivated by several important developments: there has been an explosion of experimental interest in exploiting smectic liquid crystals to reliably and robustly self-assemble patterns over macroscopic lengthscales. Applications have been driven by advances in surface control, leveraging surface patterning, topographical features such as grooves or posts, confinement in droplets or upon curved surfaces to produce emergent patterns that are optically active as lenses, gratings, photonic crystals or lithographic templates. Moreover, defect structures in the texture can be used to efficiently trap dispersed nanoparticles, making these materials useful for hierarchical or synergistic assembly processes that could potentially be adopted for metamaterial, sensor or solar cell production. Entirely new classes of layered media, such as colloidal smectics and nanocomposites, are beginning to emerge for which the classical approaches developed in thermotropic systems may not be applicable.

Responding to these exciting new applications, theorists and computationalists have begun to develop computational approaches for smectics that aim to predict structures that arise in given geometries. A central challenge is the representation of the smectic order itself, as the de Gennes complex order parameter contains redundant information, in that the imaginary part does not have a physical significance. Computational approaches built from this theory cannot simulate $+\frac{1}{2}$ defects, or combinations of defects, and tend to produce unphysical line defects. In 2014 Pevnyi, Selinger and Sluckin proposed an alternative approach, to model the density modulation $\delta\rho$ directly as a scalar field. Their approach allows for the correct defect structures, at the expense of requiring high order derivatives.

A number of very recent approaches have aimed to address some of the challenges in simulating smectics. Xia, MacLachlan, Atherton and Farrell constructed an energy functional analogous to Pevnyi's that permits $+\frac{1}{2}$ defects and is amenable to finite element simulation, and recovered focal conic domains as stationary states of this energy. Paget, Alberti, Mazza, Archer and Shendruk have proposed a complex tensor order parameter and studied the solution landscape arising from the resulting theory. Wittmann et al. have been applying Density Functional Theory to these systems. Others, including Majumdar et al., have developed other *ad hoc* energies that allow simulations of particular geometries.

In parallel with these developments, a good deal of work has been performed in the applied math community on closely related problems, and considerable expertise with techniques such as moving meshes, optimal control, deflation and a wealth of discretizations exist. The goal of the conference was to place these many efforts in communion to develop a community able to perform the modeling tasks badly needed to interpret and explain experimental observations.

3 Presentation Highlights

The program opened with a welcome and "goal-setting" session that highlighted some of the themes discussed above. Among the identified goals for the workshop was to "pin down" a set of target models and parameters, identifying challenging problems to simulate. Participants were mindful of trying to identify what has changed in this field in the last 10-20 years, and what opportunities that presents. An interesting challenge in this direction is understanding both the limitations of experiments (what we can hope to learn from them) and the complex data that is produced, and is quite distinct from the data produced by simulation. Among the challenges identified were

- Can we understand and simulate dynamic problems? What are the relevant time scales? What insights might we gain?
- The effects of multiple length scales in layered fluids, and the coupling between those scales.
- The vast number of proposed models in these areas, and the relationships between them.
- The importance of phase transitions, and understanding how to interpret models of different phases or of the transitions themselves.
- The importance of uncertainty in these models, and the role this plays in experimentation and simulation.

A meta-goal that was also identified was to discuss the potential game-changing outcomes of this field of research, answering the question *What could we do with improved understanding?*, both from the perspective of growing the community of researchers and that of securing future funding for work in this area.

Three tutorial-style presentations were given on Monday and Tuesday mornings of the workshop. First, **Randall Kamien** gave an overall tutorial "Introduction to smectics", discussing the theoretical modeling of smectic liquid crystals as a prototype layered system, including discussing several of the potential models. On Tuesday morning, **Emmanuelle Lacaze**, **Francesca Serra** and **Mohamed Gharbi** led a roundtable discussion on experimental methods, discussing what experiments were feasible, what we might learn from them, and the different experimental techniques that are common in the field. Later, **Patrick Farrell** and **Scott MacLachlan** presented a finite-element tutorial, discussing these tools in general, and details relevant to the simulation framework that has been recently proposed for these materials. All of the tutorials led to lively discussions, and clearly benefited their intended audiences.

The remaining presentations could be broadly sorted into talks on the theory, experimentation, and simulation of layer complex fluids, which were (purposefully) interspersed throughout the program. We summarize these talks below in chronological order, rather than by topic.

Jonathan Selinger presented work on different models for smectic liquid crystals, and relevant goals for a "useful" smectic theory. He reviewed the history of smectic liquid crystals, from understanding the nature of the transition in early work, to exploiting the celebrated analogy between smectics and superconductors to predict the twist-grain-boundary phase, which is an analog of the vortex lattice that arises in superconductors. Having described the successes of the Landau-de Gennes approach, he also noted some challenges for predicting the configuration of smectics under confinement that motivate the present conference.

Mohamed Gharbi showed how to experimentally control focal conic domains in smectics using substrates with undulated ridges. The ridges create (unstable) defect lines in the nematic phase, which in turn drive the formation of well-ordered arrays of smectic focal conics. In addition to this 1-d periodic array, his group created 2-d periodic arrays with alternating regions of positive and negative Gaussian curvature. Here the difference between the nematic and the smectic phase is striking: while the nematic phase has defect lines wandering around the pattern and pinned at various locations, the transition to smectics creates a perfectly ordered 2-d array of focal conic defects with high eccentricity. This can be used as a method for reconfigurable particle assembly.

Lisa Tran showed a different layered system: cholesteric liquid crystals, where the nematic director twists and the twisted director forms pseudolayers. When these liquid crystals are confined to a spherical shell, they form topological defects. By varying the composition of the aqueous medium surrounding the cell, it is possible to induce an anchoring transition from planar to homeotropic and vice versa, and observe the characteristic patterns. The transition shows a regime where a Helfrich–Hurault undulated structure appears.

Jean de Dieu Niyonzima presented a detailed X-ray study on a thin layer of smectic liquid crystals with competing boundary conditions, uniform planar at the LC-glass interface and homeotropic (perpendicular) at the LC-air interface. These boundaries are known to form oily streaks, periodic 1-d structures. The study captured the details of the smectic configuration inside the oily streaks, providing an almost complete picture of the layer structure. Remarkably, much of the bending energy of the layers is released with the formation of grain boundaries and thin isotropic boundary layers between the perpendicular and planar anchored regions.

Emmanuelle Lacaze further described the oily streaks, including the role of adding nanoparticles. The study shows that nanoparticle assembly is hierarchical: as the oily streaks contain various types of defects, nanoparticles first fill out the larger defects, then the smaller ones and only finally the defected boundary between planar and homeotropic anchoring. When assembled in defects, the absorption spectrum of the nanoparticles is altered due to changes in the plasmon resonance.

Francesca Serra presented a study of smectic focal conic domains as micro-lenses due to their refractive index gradient. The micro-lenses can be assembled in structures resembling the compound eyes of flies and they are sensitive to linear polarization of light. If a small amount of chiral dopant is added to the smectic liquid crystals, a phase transition to the cholesteric phase maintains some memory of the smectic defects as new circular defects form.

Patrick Farrell presented the work that motivated the organization of this workshop, on finite-element discretization of a Q-tensor model for smectic-A liquid crystals. He described the finite-element framework, and showed results for three "typical" simulations, one of oily streaks, one of a bend deformation, and one of focal conic domains. These are the first simulations of many smectic structures (particularly in three dimensions).

Tyler Shendruk discussed experimental observations of growing microbe communities such as E. Coli, that demonstrate local smectic order. He presented a new approach to modelling simple lamellae smectics using a complex tensor E that incorporates the degree of layering, compression deformations and orientation. He presented some numerical results that show theory constructed from this object can reproduce key topological defects identified by Selinger and Sluckin. Tyler also made some important connections to active nematic simulations and automated analysis of topological defects in dynamical simulations. He concluded by discussing possible extensions of this work to true smectic order.

René Wittman described a new and exciting class of layered material, colloidal smectics. These are phases that occur for hard extended objects, such as diskorectangles, at high aspect ratio and intermediate density. He introduced the Density Functional Theory approach, and showed some results of DFT calculations in confined geometries, contrasting them with particle simulations. These simulations show some novel defect structures between grain boundaries and René discussed their topological classification, using results from different polygonal containers as an illustration. He concluded by showing results for an annular container, showing solutions that incorporate varying numbers of radial disclination lines depending on the modulus of the annulus.

Apala Majumdar discussed her work on simulating smectic shells, using a method that incorporated a nematic Q-tensor and a scalar field ψ to describe layering. While this approach does not capture certain types of topological defects as others discussed, it is suitable for applications where they do not emerge. She provided some illustrative results, including spontaneous deformation of the layers.

Alison Ramage presented a moving-mesh finite-element method targeting enhanced resolution around defects in liquid crystals. These methods add an ”r-refinement” step to mesh adaptation, using an auxiliary error indicator to drive reallocation of mesh points to regions in the mesh where the solution is underresolved. She discussed suitable choices of monitor function, and compared the performance of several possible algorithms that could be formulated in this general approach. The numerical results for nematics appear impressive, resolving the motion of defects in a time dependent Pi-cell problem, and could be adapted for smectodynamics.

Abdalaziz Hamdan presented an extension to the finite-element framework in the work of Xia et al. that Farrell had earlier presented. This work makes use of a reformulation of the continuum equation, introducing an auxiliary variable for the gradient of the smectic density variation and a Lagrange multiplier to constrain it. While adding more variables adds some concrete cost to the discrete representation (by adding more degrees of freedom), it is a cost that can be recovered if the reformulated system can be solved using more efficient algorithmic techniques, which is the case for this approach.

Bruno Zappone focused on the transition between 1D oily streaks and 2D focal conic domains in thin films of smectic liquid crystals with homogeneous surface alignment. This transition occurs as a function of film thickness, typically when the film is between 1 and 2-μm thick. Using thin cells with controlled, varying thickness, it is possible for the two defect types to coexist. This phenomenon consolidates the analogy between the phase behavior of smectic liquid crystals and superconductors.

Teresa Lopez-Leon reviewed the progress made on the fabrication and the study of smectic liquid crystals in shells. Using microfluidics it is possible to create shells with varying thickness, whose boundary conditions (anchoring) can be controlled and tuned by changing the composition of the aqueous media inside or outside the shell and by varying the thickness of the smectic shell. A change in salt concentration of the outer media induces a shrinkage of the smectic shells that collapses into an ellipsoidal droplet, whose aspect ratio is determined by the shell's thickness.

Nigel Mottram showed a study of nematic droplets wetting a surface, a process that is fundamental in the manufacturing of liquid crystal displays. The droplet has surface tension, elasticity and anchoring and, most importantly, is anisotropic. The interplay of these elements gives rise to a very rich behavior and plays a role in the unusual wetting behavior of nematics and in some unwanted effects in the fabrication of displays.

Shawn Walker introduced optimal control approaches and their application to liquid crystal problems, inspired by the multiplicity of solutions present in configurations around Janus particles. In some experimental scenarios, it is desirable to identify the correct control responses—boundary conditions, applied fields etc.—that promote a target state and optimal control provides an approach to do so.

James Jackaman presented preliminary results on a finite-element simulation framework for smectic C liquid crystals. He highlighted that a major limiting factor in his work is the lack of existing experimental studies of the physical constants needed for these models, which prompted a lively discussion of what constants might be known, and where proposed measurements could be found.

Sean Hare presented his study of memory effect at the phase transition between smectic and cholesteric liquid crystals. Both phases have layers (or pseudolayers) but their structure is very different. At the phase transition, some memory of the topological defects is retained. An array of patterned channels with periodic indentation allows for a systematic and quantitative study of memory. The work uses an automated algorithm to detect and count focal conic domains.

Daniel Beller discussed the possibility of studying smectic order as a perturbation from the nematic phase, in effect modifying the Landau-de Gennes description of nematics. He showed an illustrative geometry: hybrid aligned nematic cooled through the $N-S_A$ transition, which settles into a network of defects near the transition and then a Toroidal Focal Conic packing in the smectic phase. He showed simulations of this geometry using the nematic theory, but allowing the twist and bend constants to diverge. He then discussed a second geometry, motivated by experiments, including hybrid anchoring on an "egg-crate" homeotropic surface where the focal conics are elliptic-hyperbolic. Beller discussed different minimization strategies to obtain equilibrium configurations, showing promising results that reproduce experimental phenomena. He also presented work on nanoparticle assembly guided by the smectic. Beller concluded with some interesting preliminary results on smectics with deformable boundaries, constructing what is, in effect, a phase field model.

Timothy Atherton presented his work on shape optimization problems, where the internal order of a liquid crystal can deform the boundary of a liquid crystal droplet or *tactoid*. He showed illustrative results using his *morpho* simulation framework for nematic tactoids as well as the deformation of a tactoid by an electric field. He discussed the application of these methods to two kinds of layered medium: First, in a cholesteric liquid crystal the shape can be commensurate with geometry, favoring spherical shapes, or incommensurate, leading to a rich variety of shapes. Second, Atherton showed preliminary results for smectic liquid crystals, using the energy presented by Apala Majumdar the day before. He concluded by calling for renewed attention to anchoring functionals for layered media, which have only been discussed so far in a very preliminary manner.

The workshop concluded with a wrap-up discussion on Friday morning, which is summarized in the discussion below.

4 Scientific Progress Made

The workshop succeeded in uniting three disparate communities—experimental physicists, theoretical physicists and applied mathematicians—who have all studied layered media or closely related problems, but have had little previous contact in the past. A considerable degree of cross-fertilization and education took place about techniques that could be relevant for those in other disciplines. For example, the physicists at the workshop felt that they came away with a better appreciation for what the finite-element simulation framework can do for them. Mathematicians and theoretical physicists gained a new appreciation of cutting-edge materials and some of the challenges in understanding them, e.g. many of the characterization techniques provide quite indirect information about the structure. Both these groups came away with new ideas about what they wanted to simulate. Experimental physicists learned that about the variety of emerging expertise in modelling layered media, and the domains of applicability of each.

A major theme was that there are many important things that remain unknown. There are multiple possible choices for smectic energy functionals, but all of them have issues with identifying constants/parameters and even reproducing basic results such as the correct order of transition between phases. In comparison to the very well understood nematic phase, this makes structure prediction in realistic scenarios very challenging. There was some discussion about inferring these parameters from experiments, including using tools of optimal control or machine learning methods. An important goal for this community is to attempt to model geometries of interest and learn from this.

A valuable outcome was a consensus that a "good" energy functional should:

- Support 1/2 defects.
- Separate smectic and nematic order.

- Ensure the layer spacing of smectic order parameter is independent of nematic order parameter.
- Allow the scientist to study phase transitions without changing the model.
- Be applicable not only to the parameter regime near the phase transition, but also deep into the smectic phase.

A further challenge is that experimentally measuring the parameters of these theories is not straightforward. There exist various methods to assess elastic constants, but sometimes these methods give inconclusive or inconsistent results based on the initial hypotheses. Anchoring is even more difficult to assess—it is convolved with elasticity, as just one challenge.

Nonetheless, the workshop highlighted how important it is for theorists to have the "right" numbers to compare their models and make quantitative predictions. Instead of specific parameter values, it may be easier to figure out ratios of parameters or identify dimensionless numbers. It may also be possible to map parameters from "nearby" theories, or adopt techniques from MD experiments to estimate missing parameters. Another idea that emerged from the meeting is to set up a database of relevant constants and parameters for the most common materials with reference to the relevant literature.

It was also suggested to set up simple model systems where we could learn some basics, and explore these with multiple simulation and experimental techniques, comparing predictions across a range of methods to gain insights.

For example, some wondered if there was a bifurcation structure analogous to the Freederickz transition that we can exploit in smectics? Thickness was raised as one possibility: as you increase the thickness, you often pass from one structure to another, leading for example to undulations in some cases. Geometry could be another means to promote this kind of transition in spherical droplets or shells, or in oily streak structures for thin films.

Other experimental considerations arose. Experimental results for nematics are often very reproducible whereas smectics are more complex. Experiments may depend on parameters and external influences that are not very easily controllable. Limits exist in the lab equipment available, but the materials are also inherently hysteristic in terms of their configuration space. Kinetics often matter in getting to certain smectic states and samples can become trapped in long-lived metastable states. Participants noted the potential of image analysis software, comparing two samples in similar states, to find similarities, rather than looking for exact reproducibility.

As a possible means of exploring the space of configurations, participants wondered how far we can get with tools like deflation and optimal control. A number of questions were posed, for example: Are there other systematic ways to explore landscapes of solutions? Is it possible to build in known symmetries? Could we "seed" defect structure? How do we connect geometric insight into structure of focal conic domains, for example, with numerical simulations?

Finally, the role of surfaces was a major theme of future work. Typically anchoring energies developed for nematics are used, such as Rapini–Papoular. The anchoring of smectics could be strongly affected by the layered structure, and might lead to much stronger orientational dependence than in the quadratic R–P model. The possibility of using Density Functional Theory—a powerful approach that has some advantages—for further work is limited by the anchoring model used, with several possible means to address this. Further work is needed in this area.

5 Outcome of the Meeting and Future Directions

There was a general consensus among attendees that further meetings would be needed to address some of the above challenges identified. Some possible venues were identified:

- The Lorentz Center `https://www.lorentzcenter.nl`. *It's essential to involve the Dutch research community.*
- Oberwolfach Center `https://www.mfo.de`. *Primarily mathematics focussed.*
- Edinburgh ICMS `https://www.icms.org.uk`.

- École de Physique Des Houches `https://www.houches-school-physics.com`
- L'Institut d'Etudes Scientifiques de Cargese. `https://iesc.universita.corsica/?lang=en` *(Corsica)*
- Institut Henri Poincaré `https://www.ihp.fr/en`
- Telluride Science Research Center `https://www.telluridescience.org`
- Aspen Center for Physics `https://www.aspenphys.org`
- Carlsberg foundation `https://www.carlsbergfondet.dk/en` *(Denmark)*
- Novo-Nordisk foundation. `https://novonordiskfonden.dk/en/` *(Denmark) Needs close connection to industrial outcomes. May be interesting to connect to lipids.*
- Institute for Pure and Applied Mathematics `https://www.ipam.ucla.edu`
- Kavli Institute for Theoretical Physics at UCSB `https://www.kitp.ucsb.edu`
- Institute for Computational and Experimental Research in Mathematics `https://icerm.brown.edu`
- Chan-Zuckerberg Biohub in San Francisco `https://www.czbiohub.org`. *Needs a strong biology focus.*

Participants also shared suggestions for possible funding opportunities that groups could apply for:

- Bilateral opportunities for collaborative funding; e.g., US/UK, Canada/France.
- Network-to-network NSF grants `https://www.nsf.gov/pubs/2021/nsf21511/nsf21511.htm`
- Simons foundation `https://www.simonsfoundation.org`
- Human Frontiers Science Program `https://www.hfsp.org/funding/hfsp-funding/research-grants` *International, people who have not collaborated before, biologically oriented but not medical.*
- Leverhulme Trust `https://www.leverhulme.ac.uk`
- ERC synergy grants `https://erc.europa.eu/apply-grant/synergy-grant` *EU based teams.*
- NSF Focused research groups `https://www.nsf.gov/mps/dmr/awards/frgs.jsp` *Need to be US based.*
- Novo nordisk New Exploratory Research and Discovery program. `https://novonordiskfonden.dk/en/grant/new-exploratory-research-and-discovery-nerd-2023/`
- A number of participants mentioned various micro-funding opportunities for short visits, hosting sabbaticals, student visits, etc.

Participants also shares some open questions and thoughts that could lead to funding ideas:

- Could we broaden the community by bringing in people looking at other kinds of layering? For example, expertise exists in the phase-field crystals communities that could be connected. What gaps exist in modelling, simulation, and experiments in other layered media?
- Why are there so many layered biological systems and yet focal conics are not observed in these systems? Is it a question of thickness? More generally, what are the growth processes in biological layered media?

- From a math perspective, layered media are far beyond simple models of fluid flow like Poisson or Stokes. Mathematicians present were excited by the diversity of physical systems to gain insight into. Possible applications raised by participants include superhydrophobic surfaces, microlenses and solar cell manufacturers.

Other possible targets of future work included:

- Community work towards gathering lists of parameters and models, for example a community database or digital hub. This could become a deliverable on a grant proposal for example.
- A review article or op-ed for liquid crystals discussing the recent developments highlighted by the conference, and advocating for areas of future work.