Chapter 1

Introduction

Liquid crystals are beautiful and mysterious. P. G. de Gennes

Currently you might be reading this work on a computer screen which is composed of millions of small cells filled with a liquid crystal. When you move the mouse, the computer sends orders, via electrical impulses, to recompose the liquid crystal structure inside the cells quickly which results in the familiar smooth scroll of the pointer on your screen. The use of these materials in laptops, digital cameras, calculators, watches etc. has made them very common in our lives. However, what is a liquid crystal at all? As its own name points out, the liquid crystalline phase is a *mesophase* between the liquid and solid phases. This means, that the molecules which constitute the material are more ordered in this phase than in the liquids, but less than in the solids. This is a wide definition, and indeed many kinds of liquid crystal phases can be found depending on their internal order.

The least ordered liquid crystal phase is the nematic N phase. Here, the molecules do not exhibit any positional order, but they possess a certain degree of orientational order, i.e., the molecules can move as freely as in a liquid but they tend to point to some direction. This direction is specified by the director \vec{n} . These materials are the most common ones for applications nowadays and have been intensively investigated during the last decades. Recently, cholesteric (or chiral nematic) liquid crystals have also become of great interest. A cholesteric mesophase or N^{*}-phase is the chiral version of the nematic phase N. It forms a spontaneous helical superstructure with a

twist axis perpendicular to the director. The structure is periodic along the helical axis with a period of half the pitch, where the pitch is the distance over which the director completes a full 360° rotation.

The director configuration in a sample of a confined liquid crystal, subjected to external fields and boundary conditions, forms a well determined structure. This ordering of the molecules induces an anisotropy in the mechanical, optical, magnetic and electrical properties on the macroscopic level. Due to this anisotropy, when an external field is applied to the sample, its director configuration and consequently its optical properties are strongly modified. This is the basis of most applications in which the properties of light, reflected or refracted by a confined liquid crystal, are changed arbitrarily by applying a voltage.

Liquid crystals can be investigated in a similar way. The anisotropic optical properties of the material allow for characterization of the director deformations by analysing the transmitted or reflected light. The most frequently applied optical method is the shadowgraph method which consists in "taking a photo" of the sample by using polarizing microscopy. Other procedures like interferometry are also very useful.

Director structures in liquid crystals were widely investigated in the past. In consequence, a basic physical description was established. The static elastic free energy was proposed by Frank (1958) and the coupling between the director orientation and flow was described macroscopically by Ericksen (1966), Leslie (1968) and Parodi (1970). During the following years, the complexity and richness of liquid crystals were appreciated, since different structures required very different methods for its investigation. A comprehensive review of the liquid crystal physics can be found in de Gennes & Prost (1993) or Chandrasekhar (1994). Additionally, the main electrooptic effects are described in Blinov & Chigrinov (1994). The investigations showed that liquid crystals are not only interesting because of their applications, but also for the investigation of basic physical phenomena. They share universal features with other systems. This universality is, for example, presented in the nematic phase when studying defects or pattern formation under non-equilibrium conditions. This is particularly interesting because liquid crystals permit much simpler experiments than the ones performed in equivalent systems.

The great variety of observable phenomena and applications explains why the study of liquid crystals has gained the attention of many physicists until today. Nowadays, new exotic phases like banana phases (Pelzl et al. 1999) or blue phases (Wright & Mermin 1989) and new materials composed of liquid crystals mixed with polymers (Kundler & Finkelmann 1995) show promising properties for potential future applications. Nevertheless, the older problems are far from being resolved. Computer simulations performed by many groups as Svenšek & Žumer (2002b) allow the visibility of hidden effects (backflow) in simple configurations. Buka et al. (2002) have recently experimentally observed convection patterns for unusual parameter combinations. Furthermore, liquid crystals have proven to be a very convenient model system to investigate universal aspects of pattern formation.

The aim of this work is to throw a little more light on the liquid crystal world. In chapter 2 we present a short introduction to the study of the standard patterns which appear in a planar cell filled with a nematic liquid crystal under an electric field. Our experimental set–up is discussed in chapter 3. In chapter 4 the dynamic behaviour of defects is compared to simulations in a particular case. A new convection pattern is presented in chapter 5. The switch response of a cholesteric liquid crystal gel is examined in chapter 6. We present our conclusions at the end of this work in chapter 7. Since a variety of different topics have been investigated, we give a short introduction to each of them in the rest of this section.

Defects

The understanding of defects is an old problem in liquid crystals. In the early beginning of the liquid crystal investigation, Friedel (1922) coined the name for the nematic phase from the Greek word "nemato" (thread) owing to the thread–like formations that the defects form in this kind of liquid crystal. Indeed, many phases are identified by observing defects on the sample, so that they can be considered as a "label" of liquid crystals. A review of possible textures which are used for identifying a liquid crystal can be found in Dierking (2003). Defects play an important role in liquid crystals: they are decisive in the phase transitions; they crucially affect some physical properties and are important for various applications. In addition, since most defect properties, their understanding goes beyond the field of the liquid crystals and can be used to explain quite different systems. For example, Chuang et al. (1991) studied a model for cosmic string evolution using the properties of defects in the nematic phase.

Defects in a nematic phase are divided into two groups: disclinations and walls. A disclination can be naively defined as a discontinuity in the director field. Point, line or sheet disclinations can be found depending on their dimension. Due to this singularity, the standard director description is not sufficient any more and the complete tensorial order parameter theory has to be considered. The first solution for the tensorial defect core was proposed by Schopohl & Sluckin (1987). This theory introduces a very small length scale compared to the typical coherence length, which appears as a great problem for computer simulations. As a result, situations, in which disclination lines are involved, have been only recently properly simulated. The first static simulation using the tensorial description was performed by Sonnet et al. (1995). Later, dynamical problems were simulated as by Tóth et al. (2002). The walls, on the other hand, are the regions where the director changes continuously its orientation between two symmetric equivalent configurations. These equivalent configurations appear when an electric or magnetic field is applied under certain boundary conditions, and can be traced back to the invariance of the nematic phase against the transformation $\vec{n} \Leftrightarrow -\vec{n}$. In principle the description of the walls is much easier than the one of disclinations.

R. B. Meyer¹ and Stieb et al. (1975) observed that a wall can be replaced by two disclination lines of strength $\pm 1/2$ at high fields. This structural transition is strongly discontinuous, i.e., a high free energy barrier exists between the two structures. When increasing the field, the two disclination lines appear in a nucleation point that propagates through the wall. New techniques for simulations and for analysis of images permit a new approach to the dynamical behaviour of this system as it is shown in chapter 4. There, the experimental results are compared with numerical simulations and with a bifurcation diagram which illustrates the process.

Pattern formation under non-equilibrium conditions

Convection in fluids is the classical example for the study of pattern formation under non-equilibrium conditions, due to the analogies to numerous other pattern-forming phenomena in nature. The most prominent phenomenon is the Rayleigh-Bénard convection in simple fluids, where a horizontal fluid layer is heated from below. If the temperature difference across the layer exceeds a certain threshold value, the system undergoes a bifurcation from a uniform and motionless state to a convective state. Convection may occur in form of regular patterns, e.g. stripes, squares or spirals, but also in form of a chaotic motion. Cross & Hohenberg (1993) offer a comprehensive review of non-equilibrium pattern formation. For a discussion of the latest results in

¹see de Gennes & Prost (1993).

Rayleigh–Bénard convection, see Bodenschatz et al. (2000).

Another system that has been investigated intensively both theoretically and experimentally in the context of symmetry–breaking instabilities is a nematic liquid crystal under the influence of an alternating electric field. The typical primary pattern consists of normal rolls similar to the ones observed in the case of Rayleigh–Bénard convection. Nevertheless, depending on the frequency and amplitude of the applied field in connection with the size and sign of certain material parameters, a huge variety of different convective states may arise. In Buka & Kramer (1996) the most important patterns in liquid crystals are examined using linear and nonlinear theory; for a first introduction to the problem we recommend Rehberg et al. (1989).

The primary instabilities, showing a high degree of universality, have been completely classified (Cross & Hohenberg 1993). The full classification of the secondary instabilities destabilizing the primary patterns, however, seems a tremendous task, except for the quasi-one dimensional case which was performed by Coullet & Iooss (1990) using symmetry arguments. The secondary instabilities have been analysed in detail for the Rayleigh–Bénard convection in simple fluids by Busse (1978). A systematic study of the nonlinear behaviour of nematic liquid crystals is highly desirable. In chapter 5 a novel instability leading to the "corkscrew" pattern is investigated. It is manifested as a regular modulation of a wall affected by convection.

Cholesteric liquid crystal gels

To conclude, let us come back to the applications. Today's liquid crystal displays are based on the alignment of a nematic liquid crystal with the field in a twisted nematic cell. The switching process is reasonably fast. However, it relies on the usage of polarisers which reduce the transmitted light and the viewing angle. The investigations for avoiding these drawbacks have shown that cholesteric liquid crystals are good candidates for electrooptically active elements. The basis of their behaviour is the same as in the nematic cells: changes in the director structure, via an applied field, are translated into variations of the optical properties. A review of the display researches with cholesteric liquid crystal is shown in Crawford & Žumer (1996). The structures presented in this phase allow for reflective or transmissive displays which do not need any polarisers and may have two stable states at zero field. The advantage of reflective devices is obvious, when we compare the pleasure of reading a book in the old format (reflective) or on a computer screen (transmissive).

However, these materials have the disadvantage of a strong dependence on temperature and pressure. This can be solved by mixing the liquid crystal with less than 10% of a polymer forming a liquid crystalline gel. The typical polymer forms a covalently cross-linked network which stabilizes the liquid crystal structures but which also increases the switching voltages strongly. Remarkable is the reflective display constructed by Yang et al. (1994), which works between two of the characteristic structures that may occur in a cholesteric liquid crystal cell: planar and fingerprint. Recently Janssen et al. (2001) and Moriyama et al. (2003) presented displays based on a thermo-reversible (physical) cholesteric gel. Here, the mixture is gelified by a physical network (not cross-linked), which is, in principle, weaker than the covalent one. Therefore, the resulting displays may operate at lower voltage. In chapter 6, a thermo-reversible cholesteric gel consisting of a nematic liquid crystal mixed with a small quantity of a chiral gelator is investigated. The switching behaviour of this system is studied, focusing the attention on the planar-fingerprint transition. The aim of this work is not only to characterize the properties which are important for applications but also the ones which can help to describe this transition physically.