Liquid Crystalline Blue Phases and Swimmer Hydrodynamics

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Department of Physics, The Rudolf Peierls Centre for Theoretical Physics University of Oxford

Thesis submitted in partial fulfilment of the requirements for the degree of Doctor of Philosophy in the University of Oxford

 \cdot Trinity Term, 2008 \cdot

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Abstract

Soft condensed matter is concerned with the wide variety of materials, ranging from polymers to foams, that are neither crystalline, nor gaseous, but lie somewhere in between. In this thesis, we shall describe two such "soft" systems: liquid crystalline blue phases and the hydrodynamics of microscopic swimming creatures.

Blue phases epitomise the term liquid crystal, possessing three-dimensional crystalline orientational order whilst maintaining full fluidity. We describe some of their unique properties and the subtle balance between local order and topology that is responsible for their stability. The application of an electric field leads to both a distortion of the blue phase and to textural transitions, both of which we simulate numerically in good agreement with experiments, reproducing the blue phase X transition observed in the 1980s, but not identified theoretically. Finally, we show that blue phases extend beyond their traditional chiral setting and also occur in achiral nematics under strong flexoelectric coupling.

At the small length scales relevant to microscopic bacteria viscous stresses dominates over the effects of inertia and propulsion is achieved by quite different means to macroscopic organisms. We describe in detail the hydrodynamics of a simple model linked-sphere swimmer, with an emphasis on the interactions between swimmers. Time reversal symmetry is important, constraining both the locomotion of individuals and the nature of their interactions. We take this up with two examples: the hydrodynamic scattering of two swimmers, related to each other by time reversal, is shown to itself be time reversal covariant; the initial configuration being recovered exactly in the final state. Finally, a model of simple dumb-bells is described. Each swimmer is time reversal invariant and does not swim, but collective locomotion is still possible through hydrodynamic interactions, leading to a minimal description of active apolar fluids.

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PUBLICATIONS

Part of the work presented in this thesis has appeared in the following peer reviewed publications:

- G. P. Alexander and J. M. Yeomans, "Stabilizing the blue phases", *Phys. Rev. E* 74, 061706 (2006).
- G. P. Alexander and J. M. Yeomans, "Flexoelectric blue phases", *Phys. Rev. Lett.* **99**, 067801 (2007).
- C. M. Pooley, G. P. Alexander, and J. M. Yeomans, "Hydrodynamic interactions between two swimmers at low Reynolds number", *Phys. Rev. Lett.* **99**, 228103 (2007).
- G. P. Alexander and D. Marenduzzo, "Cubic blue phases in electric fields", *Europhys. Lett.* **81**, 66004 (2008).
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Further work contributing to this thesis has been submitted for publication:

- G. P. Alexander, C. M. Pooley, and J. M. Yeomans, "Scattering of low Reynolds number swimmers".
- I. Dierking, E. Lark, J. Healey, G. P. Alexander, and J. M. Yeomans, "Anisotropy in the annihilation dynamics of topological defects in nematic liquid crystals".

CHAPTER 1

Introduction to Liquid Crystals

Liquid crystals occupy an intermediate position in the traditional classification of materials as solids, liquids and gases. They possess characteristics of both fluids and solids, simultaneously exhibiting flow like a liquid and elasticity like a solid [1–5]. Typical liquid crystalline materials such as 4-pentyl-4'-cyanobiphenyl (5CB), *p*-azoxyanisole (PAA) and N-(*p*methoxybenzylidene)-*p*'-butylaniline (MBBA) are composed of long, thin, rod-like molecules. At high temperatures, or low densities, both the positions and orientations of the molecules are randomly distributed, with only short range correlations between them. At very low temperatures, or high densities, the molecules arrange themselves in an orderly fashion, occupying the sites of a lattice and all oriented along a common preferred direction. Liquid crystals exist in an intermediate regime in which the molecules adopt a preferred direction, but have no long range correlations in their positions. In continuum theories of liquid crystals this orientational order is described by a vector field **n** called the *director*, which represents a coarse grained average of the local molecular alignment [1–5].

When polarised light passes through a cell of liquid crystal it emerges unaltered only if the axis of polarisation is everywhere either parallel or perpendicular to the director. Otherwise the axis of polarisation will rotate so as to preferentially follow the local orientation of the liquid crystal. If the cell is placed between crossed polarisers then light will, or will not, pass through depending on the orientation of the liquid crystal within the cell. This is the principle behind the beautiful Schlieren textures [1, 6] (Fig. 1.1), the light and dark regions reflecting the underlying orientational order of the liquid crystal. A typical Schlieren texture displays a network of points connected by dark threads, immersed in a bright background. Two distinct types of points may be discerned: those connecting two brushes and those connecting four.



Figure 1.1: An exemplary Schlieren texture in a nematic liquid crystal. Points with two and four brushes correspond to disclinations with topological strength s = 1/2 and s = 1, respectively. Reproduced with the kind permission of Ingo Dierking.

The points themselves correspond to singularities, or defects, in the orientation field where the director is undefined. The local orientational order in the vicinity of these two types of singularity is quite different and reveals a very important feature of liquid crystalline order. Namely, the orientational order in a local neighbourhood encircling a two-brush singular point is unorientable. Thus the director is *apolar* and should be described by a *line field*, instead of a vector field, as is the case for a magnet.

However, to avoid undue mathematical complexity, and following the usual conventions in the literature [1–5], we shall continue to treat the director as a vector field and impose, by hand, the equivalence relation $\mathbf{n} \sim -\mathbf{n}$ coming from its apolar character. Just as the orientation of the director has no physical significance neither does its magnitude convey any information. Because of this, the director is always normalised to unit magnitude, $|\mathbf{n}| = 1$.

1.1 Elastic distortions

The orientational order in liquid crystals represents the breaking of a continuous symmetry. As a consequence, global rotations of the director do not cost any energy, but local rotations, leading to gradients in the director field, do [3]. There are three independent distortions of the director field in a nematic liquid crystal, known as *splay*, *twist* and *bend* [1–5] and shown in Fig. 1.2. In terms of derivatives of the director field these three distortions correspond to the



Figure 1.2: Schematic illustration of the elastic distortions in a nematic liquid crystal.

following expressions being non-zero:

$$\mathbf{n}(\nabla \cdot \mathbf{n})$$
 splay, (1.1)

$$\mathbf{n} \cdot \nabla \times \mathbf{n}$$
 twist, (1.2)

$$-(\mathbf{n} \cdot \nabla)\mathbf{n}$$
 bend. (1.3)

We adopt a slightly unconventional notation here, writing splay as $\mathbf{n}(\nabla \cdot \mathbf{n})$ instead of simply $\nabla \cdot \mathbf{n}$ and bend as $-(\mathbf{n} \cdot \nabla)\mathbf{n}$ instead of $\mathbf{n} \times \nabla \times \mathbf{n}$ as is more common in the literature [1]. The latter change is merely a matter of personal preference since the two expressions are equivalent because the director is a unit vector. We choose to write splay as $\mathbf{n}(\nabla \cdot \mathbf{n})$ both to emphasise the requirement that properties of the liquid crystal need to be invariant under $\mathbf{n} \to -\mathbf{n}$ and to reveal the vectorial character of splay distortions, an aspect of some importance when we discuss flexoelectricity in Chapter 4.

The preferred state of a liquid crystal is one of uniform alignment and therefore we can associate a free energy cost with each of these distortions, leading to the Frank free energy density [1–5]

$$\mathcal{F}_{\text{Frank}} = \frac{1}{2} K_1 \left(\nabla \cdot \mathbf{n} \right)^2 + \frac{1}{2} K_2 \left(\mathbf{n} \cdot \nabla \times \mathbf{n} + q_0 \right)^2 + \frac{1}{2} K_3 \left((\mathbf{n} \cdot \nabla) \mathbf{n} \right)^2, \qquad (1.4)$$

where K_1, K_2, K_3 are the Frank splay, twist and bend elastic constants, respectively. The constant q_0 appearing in the twist invariant is permitted on symmetry grounds only in systems lacking inversion symmetry, i.e., in chiral liquid crystals. In such chiral systems this coefficient will be generically non-zero and serves to promote a naturally distorted texture in which the director displays a pure twist distortion with $\mathbf{n} \cdot \nabla \times \mathbf{n} = -q_0$. It may readily be verified that the director field $\mathbf{n} = (0, \cos(q_0 x), \sin(q_0 x))$ satisfies this relation. It describes a helical texture



Figure 1.3: Schematic illustration of topological defects in nematic liquid crystals. (a) s = 1/2, (b) s = -1/2, (c) s = 1, splay-type ($\psi = 0$), and (d) s = 1, bend-type ($\psi = \pi/2$). In each case the director field is confined to the plane of the page and the location of the disclination line is indicated by a small black dot.

known as the *cholesteric* phase in which the director rotates moving along the helical axis, but is uniform perpendicular to it. The director undergoes a complete revolution over a distance of $2\pi/q_0$, but because of its apolar character the physical repeat length, known as the *pitch*, is precisely half this, $p = \pi/q_0$ [1].

1.2 Disclinations

Topological defects in liquid crystals give rise to both the beautiful Schlieren textures of Fig. 1.1 and the name nematic, which originates from the Greek $\nu\eta\mu\alpha$ meaning 'thread-like'. They are singular lines where the director field is undefined, and are classified by a half-integer s, which represents the winding number, or topological strength, of the disclination line. A schematic illustration of the local director field configuration for a number of different strength disclinations is shown in Fig. 1.3. The $s = \pm 1/2$ ((a) and (b)) and $s = \pm 1$ ((c) and (d)) disclinations have a fundamentally different character. Namely, only the former are topologically protected: the $s = \pm 1$ disclinations can be continuously deformed into a uniform texture and removed by purely local manipulations. By contrast, no such operations can be performed on the $s = \pm 1/2$ disclinations, which can only be removed by the annihilation of a pair of such defects of oppoA feature of some importance in the description of nematic liquid crystals is that the Frank theory assigns infinite energy to these disclination lines. In the one elastic constant approximation, $K_1 = K_2 = K_3 = K$, the director field in the vicinity of a strength *s* disclination takes the form $\mathbf{n} = (\cos(s\phi + \psi), \sin(s\phi + \psi), 0)$, where ϕ is the usual polar angle and ψ is an arbitrary constant. The energy of this field configuration is $\pi K s^2 \ln(R/r_c)$ per unit length of the disclination line, where r_c is a short distance cut-off representing the size of the defect core, typically of molecular length scales, and *R* is a large scale cut-off of order the distance to the nearest boundary or another disclination [1].

The infinite energy cost of $s = \pm 1$ disclinations can be avoided by allowing the director to 'escape in the third dimension', becoming perpendicular to the page in Fig. 1.3 at the centre of the defect [7], a situation which is observed experimentally in liquid crystals confined to cylindrical capillaries [1]. If the boundary conditions are homogeneous at the surface of the capillary, $\mathbf{n} = \mathbf{e}_{\phi}$ at $\rho = R$, and escaped along the axis, $\mathbf{n} = \mathbf{e}_z$ at $\rho = 0$, then the director field is given by $\mathbf{n} = \cos(\theta)\mathbf{e}_z + \sin(\theta)\mathbf{e}_{\phi}$, with $\theta = 2 \arctan(\rho/R)$, and the energy of the configuration is $3\pi K$ per unit length of the cylinder. This configuration, generated by a compromise between geometry and boundary conditions, displays a local twist of the director field as one moves along any straight line passing through the cylinder axis. For this reason it is reminiscent of the double twist cylinders that characterise the cholesteric blue phases and which shall be the principal focus of Chapters 2 - 4. However, the blue phases differ in a fundamental respect: there is no capillary tube anchoring the liquid crystal at its surface. Thus in the blue phases one needs both a different mechanism by which to stabilise the locally double twisted configuration, and to consider how the local texture extends to the surrounding liquid crystal in the region beyond $\rho = R$. It turns out that it is not possible to do the latter without introducing disclinations in the director field as we shall describe in § 1.5.

Finally, we mention that exactly the same comments can be made for a capillary with homeotropic anchoring, $\mathbf{n} = \mathbf{e}_{\rho}$ at $\rho = R$, leading to a locally *double splayed* cylinder. The prospect that this local configuration might also appear in an unconfined liquid crystal has been considered only relatively recently [8,9] and will be taken up in Chapter 4.

1.3 Isotropic-nematic transition

An important consequence of the nature of the local symmetry of liquid crystals is that the director, being apolar and of fixed (unit) magnitude, cannot serve as an order parameter for the isotropic-nematic transition. When a nematic liquid crystal is cooled through the isotropic-nematic transition and viewed through crossed polarisers the sample remains dark in the isotropic phase, but becomes bright following the transition to the nematic. Thus the optical response of the liquid crystal can be taken as an order parameter for the transition. In a continuous medium the optical response is determined by the dielctric tensor ϵ . This tensor may be split into two pieces, an isotropic part and a deviatoric part, just as can be done for the stress tensor in a Newtonian fluid,

$$\epsilon_{\alpha\beta} = \frac{1}{3}\operatorname{tr}(\boldsymbol{\epsilon})\delta_{\alpha\beta} + \left[\frac{1}{2}(\epsilon_{\alpha\beta} + \epsilon_{\beta\alpha}) - \frac{1}{3}\operatorname{tr}(\boldsymbol{\epsilon})\delta_{\alpha\beta}\right].$$
 (1.5)

The isotropic part is present even in the high temperature phase and does not impart any rotation to the axis of polarisation of light passing through the material. Thus the transmission of light through crossed polarisers arises solely from the non-vanishing of the deviatoric part of the dielectric tensor, which we adopt as our order parameter for the isotropic-nematic transition and denote by the symbol \mathbf{Q} . As is usual in Landau theories, the physical origin of the order parameter is of less direct relevance than its symmetry properties. For example, we could just as easily have defined our order parameter in terms of the deviatoric part of the magnetic susceptibility tensor $\boldsymbol{\chi}$ [1], and although \mathbf{Q} would then have a different physical origin, it would still have the same symmetries.

For many of our purposes it is therefore only important to note that \mathbf{Q} is a traceless, symmetric, second rank tensor. A coordinate system can always be found for such a tensor in which it is diagonal

$$\mathbf{Q} = \text{diag}\left(S, -(S+\eta)/2, -(S-\eta)/2\right).$$
(1.6)

When $\eta = 0$ the tensor exhibits *uniaxial* symmetry, which is often assumed to be the case in nematic liquid crystals. More generally, for $\eta \neq 0$, the tensor is *biaxial*, with η measuring the degree of biaxiality. This biaxiality, which can be important in the description of disclinations [10] or chiral systems [11], is not captured by the Frank director theory of liquid crystals. A Landau theory of the isotropic-nematic transition was first given by de Gennes [1]. The bulk free energy density is expanded in powers of the independent invariants of the order parameter, which we take to be $tr(\mathbf{Q}^2)$ and $tr(\mathbf{Q}^3)$. The simplest expression for the bulk free energy density is then

$$\mathcal{F}_{\text{bulk}} = a \operatorname{tr}(\mathbf{Q}^2) - b \operatorname{tr}(\mathbf{Q}^3) + c \left(\operatorname{tr}(\mathbf{Q}^2)\right)^2.$$
(1.7)

The existence of a cubic term in the free energy density indicates that the transition will be first order, as observed experimentally. The first order nature of the transition raises the question of whether it is appropriate to truncate the expansion at fourth order, as done here, or also retain terms up to sixth order. In a mean field calculation, one finds that the fourth order truncation always yields a first order transition with a uniaxial \mathbf{Q} in the nematic phase. If, however, terms up to sixth order are retained, then not only can the transition lead to a biaxial nematic, but also there can be second order transitions between two distinct uniaxial nematics in the ordered phase [1].

1.4 Hydrodynamics

One of the most fascinating aspects of liquid crystals is that they flow. And while the motions of Newtonian fluids are already extremely rich and complex [12–14], the delicate coupling between fluid flow and orientational order produces a wealth of new phenomena in liquid crystals [1,15]. In general, flow leads to a rotation of the local orientation, which in turn influences the flow. Similarly, if a disturbance is initiated in the director, its reorientation is generally accompanied by fluid motion, an effect sometimes referred to as *backflow*.

The hydrodynamics of liquid crystals was developed during the 1960s, principally in the work of Ericksen and Leslie [15], although there was considerable debate over the precise form of the equations for some time [1, 16, 17]. This formulation, known as the *Ericksen-Leslie* equations, is based on the dynamics of the nematic director \mathbf{n} and is the most commonly used description of liquid crystal hydrodynamics. Only the orientational order is treated as a hydrodynamic variable, the magnitude of the order being considered fixed and homogeneous.

In systems containing defects, or close to a phase transition, however, the magnitude is not constant and can play a significant role in the hydrodynamics [18–23]. We account for this by using a hydrodynamic description based on the **Q**-tensor developed by Edwards, Beris and Grmela [24–28] and later extended into a general framework for the description of the hydrodynamics of complex fluids by Grmela and Öttinger [29–31].

The order parameter evolves according to a Ginzburg-Landau type equation describing relaxation towards the minimum of the free energy, but with a convective time derivative to account for the advection with the fluid

$$D_t \mathbf{Q} = \Gamma \left(\frac{-\delta F}{\delta \mathbf{Q}} + \frac{1}{3} \operatorname{tr} \left(\frac{\delta F}{\delta \mathbf{Q}} \right) \mathbf{I} \right).$$
(1.8)

The term in brackets on the right hand side is called the molecular field, \mathbf{H} , and Γ is a collective rotational diffusion constant. The material derivative for rod-like molecules is given by

$$D_t \mathbf{Q} = \left(\partial_t + \mathbf{u} \cdot \nabla\right) \mathbf{Q} - \left(\xi \mathbf{D} + \mathbf{\Omega}\right) \left(\mathbf{Q} + \frac{1}{3}\mathbf{I}\right) - \left(\mathbf{Q} + \frac{1}{3}\mathbf{I}\right) \left(\xi \mathbf{D} - \mathbf{\Omega}\right) + 2\xi \left(\mathbf{Q} + \frac{1}{3}\mathbf{I}\right) \operatorname{tr} \left(\mathbf{Q}\mathbf{W}\right),$$
(1.9)

where $\mathbf{D} = (\mathbf{W} + \mathbf{W}^T)/2$ and $\mathbf{\Omega} = (\mathbf{W} - \mathbf{W}^T)/2$ are the symmetric and antisymmetric parts, respectively, of the velocity gradient tensor $W_{\alpha\beta} = \nabla_{\beta} u_{\alpha}$. The constant ξ depends on the molecular details of a given liquid crystal.

The fluid velocity field is taken to obey the continuity equation and a Navier-Stokes equation with a stress tensor generalised to describe liquid crystal hydrodynamics

$$\partial_t \varrho + \nabla \cdot \left(\varrho \mathbf{u} \right) = 0 , \qquad (1.10)$$

$$\varrho(\partial_t \mathbf{u} + \mathbf{u} \cdot \nabla \mathbf{u}) = -\nabla p + \nabla \cdot \boldsymbol{\sigma} , \qquad (1.11)$$

$$\sigma_{\alpha\beta} = \mu \left(\nabla_{\alpha} u_{\beta} + \nabla_{\beta} u_{\alpha} \right) + 2\xi \left(Q_{\alpha\beta} + \frac{1}{3} \delta_{\alpha\beta} \right) Q_{\gamma\delta} H_{\gamma\delta} - \xi H_{\alpha\gamma} \left(Q_{\gamma\beta} + \frac{1}{3} \delta_{\gamma\beta} \right) - \xi \left(Q_{\alpha\gamma} + \frac{1}{3} \delta_{\alpha\gamma} \right) H_{\gamma\beta} + Q_{\alpha\gamma} H_{\gamma\beta} - H_{\alpha\gamma} Q_{\gamma\beta} - \nabla_{\alpha} Q_{\gamma\delta} \frac{\delta F}{\delta \nabla_{\beta} Q_{\gamma\delta}} .$$

$$(1.12)$$

Eqs. (1.8) - (1.12) are collectively referred to as the *Beris-Edwards* equations and we shall use them throughout this thesis to describe the time evolution of the **Q**-tensor. To solve these equations we employ a standard finite difference scheme for the evolution of **Q**, Eq. (1.8), and a three-dimensional lattice Boltzmann algorithm [32–34] for the fluid, Eqs. (1.10) and (1.11).



Figure 1.4: (a) Typical texture of blue phase I under reflecting microscopy taken from Reference [50]. (b) A single monodomain of blue phase I exhibiting facetting and growth in a sequence of steps, from Reference [35].

1.5 Blue phases: a pictorial introduction

Blue phases are found in highly chiral liquid crystals in a narrow temperature window (typically < 1 K) between the high temperature isotropic fluid and low temperature cholesteric phases. They are remarkable mesophases, exhibiting a brightly coloured texture of individual, micron sized platelets, as shown in Fig. 1.4. The bright colour indicates selective reflection due to a periodic structure, much like in an ordinary crystal, but with a much larger characteristic length scale, and indeed the reflection spectra show Bragg peaks that can be indexed by cubic space groups with lattice constants of several hundred nanometers. Furthermore, individual platelets of monodomain crystals themselves show distinctive facetting corresponding to Miller planes of the lattice structure and with the faces growing in a sequence of steps [35]. And yet, blue phases are not crystals in the traditional sense: they have *no* long range positional order and are full three-dimensional fluids [11]. The crystalline order is in the orientational degrees of freedom of the liquid crystal.

The key to understanding their unique properties was in realising that the locally preferred ordering in a chiral liquid crystal is one of *double twist* as opposed to the usual single twist of the ordinary cholesteric helix [11,36–38]. This local texture of the blue phases can be thought of as pieces of cholesteric helix simply added together. As an example, consider superposing three pieces of cholesteric with their helical axes rotated from one another by 120°. As illustrated in Fig. 1.5 these three pieces combine to produce a local double twist, but in addition, because of the way they were chosen, they also impose hexagonal symmetry. This hexagonal region



Figure 1.5: A pictorial construction of the planar hexagonal blue phase. Pieces of three cholesteric helices, with their axes rotated from one another by 120° are superposed to form a region of local double twist, which may then be used as a unit cell for a two dimensional hexagonal lattice. But is the correct unit cell (a) or (b)?

may then be adopted as a unit cell and used to generate a two-dimensional lattice. However, it is clear that this is not an innocuous process that smoothly extends the local order globally. Rather, the vertices of the unit cell boundary generate a lattice of s = -1/2 disclination lines, so that the texture is in this sense frustrated.

The construction we have just described encapsulates the generic features of crystalline blue phases: they are composed of an array of energetically favourable *double twist cylinders* separated by a *network of disclinations*. This structure also explains why the blue phases are stable only in a narrow temperature window at the isotropic-cholesteric phase boundary, as here the magnitude of the order is small and hence the free energy penalty associated with disclinations is low.

Three distinct blue phase textures have been identified experimentally, separated by first order phase transitions and occuring in the order blue phase III, blue phase II, blue phase I upon cooling from the isotropic liquid [39, 40]. Exemplary phase diagrams obtained in Reference [40] are shown in Fig. 1.6. The textures that are observed experimentally, however, do not possess the two-dimensional hexagonal symmetry of our simple example. The disclinations form textures with cubic symmetry and the stable phases have space groups $O^{8-}(I4_132)$ and $O^2(P4_232)$, corresponding to blue phase I and blue phase II, respectively [41–43]. Blue phase



Figure 1.6: Experimental phase diagrams of the cholesteric blue phases for two different chiral compounds, taken from Reference [40]. Three distinct blue phases are found, in the order blue phase I, blue phase II, blue phase III upon increasing the amount of chiral dopant.

III is somewhat different and has an amorphous structure with the same symmetry as the isotropic fluid [11]. The director field texture and properties of blue phase III are not as well understood as those of the crystalline blue phases [44, 45], which will be the primary focus of our attention.

The unique combination of crystalline order with lattice constants comparable to the wavelength of visible light and full three-dimensional fluidity make the blue phases ideal for technological uses such as fast light modulators, photonic crystals or tunable lasers [46–49]. The principal obstruction to fulfilling this potential is the very limited temperature range over which blue phases have traditionally been thermodynamically stable. However, in recent years, novel techniques have extended the stability range to as much as 60 K, including room temperature, by the addition of bimesogenic molecules or photo-crosslinking of polymers [50,51]. The development of blue phase materials for technological uses has now progressed to the stage where, in May 2008, Samsung Electronics unveiled a prototype 'blue phase mode LCD' at the Society for Information Display 2008 International Symposium [52].

Many of the potential technological applications of blue phases, like traditional liquid crystal displays, rely upon the response of the material to an electric field for their operation. In the blue phases the effects of an electric field are particularly rich, including continuous distortions of the size and shape of the unit cell and a series of field induced textural transitions to new blue phase structures, not stable in zero field [11, 53]. This is a classic problem, worked on extensively during the hey-day of the blue phases in the 1980s, and yet many features remain only partially explained.

The increased stability of the new blue phase materials is not understood. Traditional descriptions of the blue phases ascribe their stability to a fine energetic balance between favourable regions of double twist and unfavourable disclination lines. In director field, or low chirality, theories the latter are thought of as having isotropic cores, which become increasingly costly as the temperature is lowered below the isotropic transition. To increase their stability one must either make the double twist regions more beneficial, or the disclinations less costly. It is natural to speculate that in the case of the polymer stabilised blue phases [51], the polymer sits preferentially in the defect core regions, whereupon photo-crosslinking effectively solidifies the texture, but this has not yet been verified.

In the opening part of this thesis we shall investigate the properties of the crystalline blue phases, presenting first a description of their structure and thermodynamic stability obtained within the Landau-de Gennes framework, and subsequently an analysis of their behaviour under an applied electric field.

We close this chapter by recalling the words of Sir Charles Frank when describing blue phases [11].

"They are totally useless, I think, except for one important intellectual use, that of providing tangible examples of topological oddities, and so helping to bring topology into the public domain of science, from being the private preserve of a few abstract mathematicians and particle theorists."

CHAPTER 2

Blue phases: the isotropic-cholesteric transition

The theoretical description of the general features and properties of the blue phases was developed in the 1980s [11,36,37,42,43]. Two approaches were followed: a low chirality theory based on the Frank director field description [36,37] and a high chirality theory using the Landau-de Gennes **Q**-tensor [42,43]. Although broad agreement was reached that the thermodynamically stable phases all had the cubic symmetry observed experimentally, the agreement with experiment remained incomplete. In particular, the O^{8-} texture identified with blue phase I was not found to undergo a direct transition to the isotropic liquid, but always appeared between blue phase II and the cholesteric phase. Furthermore, additional textures with space groups O^{8+} and O^5 were found to be stable within the parameter range corresponding to the experiments, but were not observed experimentally. Both of these discrepancies were overcome by a numerical minimisation of the Landau-de Gennes free energy [54], within the one elastic constant approximation.

However, the report by Coles and Pivnenko [50] of a blue phase with a substantially larger temperature range does not fit into the established theoretical framework and merits closer attention to understand where its increased stability originates from. In this Chapter we shall describe a numerical determination of the phase diagram of cholesteric liquid crystals, obtained for an extended Landau-de Gennes theory, and investigate the effects of varying material parameters on the blue phase stability. These include the three Frank elastic constant and an additional chiral invariant that allows for an inversion of the helical sense in the cholesteric phase. We begin with a review of the relation between the Frank director (§ 1.1) and Landaude Gennes (§ 1.3) theories, thereby establishing the extended Landau-de Gennes free energy, Eq. (2.21), that we minimise to construct the phase diagram.

2.1 Relating the Landau-de Gennes and Frank theories

The Frank director and Landau-de Gennes **Q**-tensor theories provide two different ways of describing the physical properties of liquid crystals. Each of these approaches has a different emphasis and one often complements the other. Indeed, additional insight can frequently be gained by comparing and relating the two descriptions.

The director field \mathbf{n} may be used to define a traceless, symmetric, second rank tensor

$$\mathbf{Q}_{\text{uniaxial}} = \frac{3S}{2} \left(\mathbf{n} \otimes \mathbf{n} - \frac{1}{3} \mathbf{I} \right) \,, \tag{2.1}$$

through which a relationship between the Landau-de Gennes and Frank theories may be established. The free parameter S in this mapping represents the magnitude of the liquid crystalline order and is often referred to as a scalar order parameter. For a given director field, Eq. (2.1) allows us to construct a **Q**-tensor. However, by construction this **Q**-tensor is uniaxial so that, in general, the procedure cannot be inverted to extract a director field from **Q**. To do this we instead note that the director is an eigenvector of **Q**_{uniaxial} that, provided S is positive, corresponds to the maximal eigenvalue. We will continue to use this as a definition of the director even when **Q** is not uniaxial. By this definition the director is well-defined and regular so long as the maximal eigenvalue of **Q** is unique. Conversely, if the maximal eigenvalue is degenerate then the director is undefined, indicating the presence of a disclination. Since two eigenvalues are equal, these disclinations in the director always coincide with points where the **Q**-tensor is uniaxial, a necessary and sufficient condition for which is that [55]

$$\left|\mathbf{Q}\right|^{6} = 54\left(\det(\mathbf{Q})\right)^{2}, \text{ and } \det(\mathbf{Q}) < 0.$$
 (2.2)

The Frank free energy, Eq. (1.4), describes the energy penalty associated with spatial inhomogeneity in the direction of alignment of the liquid crystal. Within the Landau-de Gennes theory this is accounted for by supplementing the bulk free energy density, Eq. (1.7), with a gradient free energy density

$$\mathcal{F}_{\text{gradient}} = \frac{L_1}{2} \left(\nabla \mathbf{Q} \right)^2 + \frac{L_2}{2} \left(\nabla \cdot \mathbf{Q} \right)^2 + 2q_0 L_1 \mathbf{Q} \cdot \nabla \times \mathbf{Q} , \qquad (2.3)$$

containing all the independent symmetry allowed invariants up to quadratic order in both

gradients and \mathbf{Q} [1]. In the cholesteric systems that will be the focus of our attention it is frequently convenient to express the gradient free energy density differently, combining the term linear in gradients with the $(\nabla \mathbf{Q})^2$ term by 'completing the square' [11]

$$\mathcal{F}_{\text{gradient}} = \frac{L_{21}}{2} \left(\nabla \times \mathbf{Q} + 2q_0 \mathbf{Q} \right)^2 + \frac{L_{22}}{2} \left(\nabla \cdot \mathbf{Q} \right)^2 \,. \tag{2.4}$$

The two expressions, Eqs. (2.3) and (2.4), are identical up to total derivatives and a tr (\mathbf{Q}^2) term, which may be thought of as 'borrowed' from the bulk free energy density. The insight gained from this rewriting of the gradient free energy density is that minimisers can be found by the solution of a first order differential equation

$$\mathbf{Q} = -(1/2q_0)\nabla \times \mathbf{Q} , \qquad (2.5)$$

instead of the usual second order Euler-Lagrange equations. The solution of Eq. (2.5),

$$\mathbf{Q} \sim \begin{pmatrix} 0 & 0 & 0 \\ 0 & \cos(2q_0 x) & \sin(2q_0 x) \\ 0 & \sin(2q_0 x) & -\cos(2q_0 x) \end{pmatrix},$$
(2.6)

is known as a *biaxial helix*. Here we have chosen the helical axis to lie along the x-direction, but this is arbitrary and any SO(3) rotation of this tensor will also solve Eq. (2.5). Furthermore, since Eq. (2.5) is linear, any linear combination of biaxial helices (with any directions for their helical axes) is also a solution. This is the basis for the pictorial construction, described in § 1.5, whereby blue phases are built up as a linear combination of cholesteric helices.

Using Eq. (2.1) it is straightforward to show that the Landau-de Gennes free energy density reproduces the Frank version, with

$$K_1 = \frac{9S^2}{4} (L_{21} + L_{22}), \quad K_2 = \frac{9S^2}{2} (L_{21}), \quad K_1 = \frac{9S^2}{4} (L_{21} + L_{22}).$$
 (2.7)

This highlights a shortcoming of the quadratic Landau-de Gennes theory, which is seen to account for only two elastic constants, as compared to the three that appear in the Frank director field theory. To obtain three distinct Frank elastic constants it is necessary to extend the Landau-de Gennes theory by including terms of cubic order in \mathbf{Q} in the gradient free energy [56–58]. If we continue to confine our attention to terms of at most quadratic order in

gradients then we can construct the following invariants

$$\epsilon_{\alpha\beta\gamma}Q_{\alpha\delta}Q_{\delta\epsilon}\nabla_{\beta}Q_{\gamma\epsilon} , \qquad (2.8)$$

$$\epsilon_{\alpha\beta\gamma}Q_{\alpha\delta}Q_{\beta\epsilon}\nabla_{\delta}Q_{\gamma\epsilon} , \qquad (2.9)$$

$$Q_{\alpha\beta}\nabla_{\alpha}Q_{\beta\gamma}\nabla_{\delta}Q_{\gamma\delta} , \qquad (2.10)$$

$$Q_{\alpha\beta}\nabla_{\alpha}Q_{\gamma\delta}\nabla_{\beta}Q_{\gamma\delta} , \qquad (2.11)$$

$$Q_{\alpha\beta}\nabla_{\alpha}Q_{\gamma\delta}\nabla_{\gamma}Q_{\beta\delta} , \qquad (2.12)$$

$$Q_{\alpha\beta}\nabla_{\gamma}Q_{\alpha\beta}\nabla_{\delta}Q_{\gamma\delta} , \qquad (2.13)$$

$$Q_{\alpha\beta}\nabla_{\gamma}Q_{\alpha\gamma}\nabla_{\delta}Q_{\beta\delta} , \qquad (2.14)$$

$$Q_{\alpha\beta}\nabla_{\gamma}Q_{\alpha\delta}\nabla_{\gamma}Q_{\beta\delta} , \qquad (2.15)$$

$$Q_{\alpha\beta}\nabla_{\gamma}Q_{\alpha\delta}\nabla_{\delta}Q_{\beta\gamma}.$$
 (2.16)

The mapping of these additional invariants to the Frank theory can be done using Eq. (2.1), leading to the relations [58]

$$K_1 = \frac{9S^2}{4} \left(L_{21} + L_{22} \right) + \frac{9S^3}{8} \left(-L_{33} - 2L_{34} - L_{35} + 2L_{37} + L_{38} + 2L_{39} \right), \qquad (2.17)$$

$$K_2 = \frac{9S^2}{2} (L_{21}) + \frac{9S^3}{8} (-2L_{34} + L_{38}) , \qquad (2.18)$$

$$K_3 = \frac{9S^2}{4} \left(L_{21} + L_{22} \right) + \frac{9S^3}{8} \left(2L_{33} + 4L_{34} + 2L_{35} - L_{37} + L_{38} - L_{39} \right), \qquad (2.19)$$

$$K_2 q_0^{\rm F} = \frac{9S^2}{2} (L_{21} q_0) + \frac{9S^3}{8} (L_{31} q_0 + L_{32} q_0) .$$
(2.20)

If all of the cubic order invariants are retained there will be many more Landau-de Gennes elastic constants than there are Frank constants and the theory will contain a large number of phenomenological parameters that it is not realistic to study the effects of in detail. Consequently we shall restrict our attention to a subset, choosing to focus on three invariants; (2.8), (2.11) and (2.15). These are chosen for a number of reasons: first, it is clear that one should retain at least one chiral and one achiral invariant. Since there does not appear to be any substantial difference between the two chiral invariants we feel confident that no qualitative changes would result from retaining both terms. It is more difficult to choose representatives of the achiral invariants. The invariant (2.15) is retained because, uniquely amongst the achiral invariants, it contributes equally to all three Frank elastic constants. This leaves only a choice of invariant that will distinguish between the Frank splay and bend elastic constants. We choose to use (2.11) partly because it has been used in previous work [18,59], partly because it gives the largest distinction between K_1 and K_3 and partly because we found that it has the largest contribution to the energetics of a single double twist cylinder. The extended version of the Landau-de Gennes free energy then takes the form

$$F = \frac{1}{V} \int_{\Omega} d^3 r \left\{ \frac{A_0(1-\gamma/3)}{2} \operatorname{tr} \left(\mathbf{Q}^2 \right) - \frac{A_0 \gamma}{3} \operatorname{tr} \left(\mathbf{Q}^3 \right) + \frac{A_0 \gamma}{4} \left(\operatorname{tr} \left(\mathbf{Q}^2 \right) \right)^2 \right. \\ \left. + \frac{L_{21}}{2} \left(\nabla \times \mathbf{Q} + 2q_0 \mathbf{Q} \right)^2 + \frac{L_{22}}{2} \left(\nabla \cdot \mathbf{Q} \right)^2 + q_0 L_{31} \mathbf{Q}^2 \cdot \nabla \times \mathbf{Q} \right.$$

$$\left. + \frac{L_{34}}{2} Q_{\alpha\beta} \nabla_{\alpha} Q_{\gamma\delta} \nabla_{\beta} Q_{\gamma\delta} + \frac{L_{38}}{2} Q_{\alpha\beta} \nabla_{\gamma} Q_{\alpha\delta} \nabla_{\gamma} Q_{\beta\delta} \right\},$$

$$(2.21)$$

where in the bulk free energy A_0 is a constant with the dimensions of an energy density and γ plays the role of an effective temperature for thermotropic liquid crystals [28, 32].

2.2 Isotropic–cholesteric transition

The description of the transition from an isotropic fluid to a chiral liquid crystal is simplified by the use of a dimensionless form of the free energy [11, 42], obtained by defining the dimensionless length scale $x = 2q_0r$ and energy scale $f = (729/16A_0\gamma)F$, and also rescaling the order parameter by $\mathbf{Q} \rightarrow (4/3\sqrt{6})\mathbf{Q}$. In terms of these dimensionless variables the free energy, Eq. (2.21), then takes the form

$$f = \frac{1}{(2q_0)^{3V}} \int_{\Omega} d^3x \left\{ \frac{\tau}{4} \operatorname{tr} \left(\mathbf{Q}^2 \right) - \sqrt{6} \operatorname{tr} \left(\mathbf{Q}^3 \right) + \left(\operatorname{tr} \left(\mathbf{Q}^2 \right) \right)^2 \right. \\ \left. + \frac{\kappa^2}{4} \left[\left(\nabla \times \mathbf{Q} + \mathbf{Q} \right)^2 + \frac{L_{22}}{L_{21}} \left(\nabla \cdot \mathbf{Q} \right)^2 + \frac{4L_{31}}{3\sqrt{6}L_{21}} \mathbf{Q}^2 \cdot \nabla \times \mathbf{Q} \right. \\ \left. + \frac{4L_{34}}{3\sqrt{6}L_{21}} Q_{\alpha\beta} \nabla_{\alpha} Q_{\gamma\delta} \nabla_{\beta} Q_{\gamma\delta} + \frac{4L_{38}}{3\sqrt{6}L_{21}} Q_{\alpha\beta} \nabla_{\gamma} Q_{\alpha\delta} \nabla_{\gamma} Q_{\beta\delta} \right] \right\},$$

$$(2.22)$$

and is seen to depend only on the two dimensionless parameters

$$\tau := \frac{9(3-\gamma)}{\gamma} , \qquad (2.23)$$

$$\kappa^2 := \frac{108q_0^2 L_{21}}{A_0 \gamma} , \qquad (2.24)$$

known as the *reduced temperature* and the *chirality* respectively, and the ratio of elastic constants.

The isotropic-cholesteric transition can be calculated in detail analytically [11, 42], which

we review briefly, emphasising the additional features arising from the extended form of the Landau-de Gennes free energy that we use here. The \mathbf{Q} -tensor describing an ideal cholesteric helix is given by [11, 42, 60]

$$\mathbf{Q} = \frac{Q_2}{\sqrt{2}} \begin{pmatrix} 0 & 0 & 0\\ 0 & \cos(kx) & \sin(kx)\\ 0 & \sin(kx) & -\cos(kx) \end{pmatrix} - \frac{Q_h}{\sqrt{6}} \begin{pmatrix} 2 & 0 & 0\\ 0 & -1 & 0\\ 0 & 0 & -1 \end{pmatrix} , \qquad (2.25)$$

where we have taken the helical axis to define the x-direction of a Cartesian coordinate system. A short calculation shows that the free energy is

$$f = \frac{1}{4}(\tau + \kappa^2)(Q_h^2 + Q_2^2) + (Q_h^3 - 3Q_hQ_2^2) + (Q_h^2 + Q_2^2)^2 + \frac{\kappa^2}{4}Q_2^2 \left[(1 - \beta Q_h)k^2 - 2(1 - \alpha Q_h)k\right], \quad (2.26)$$

where we have defined the parameters $\alpha := -2L_{31}/(9L_{21})$ and $\beta := 2(2L_{34} - L_{38})/(9L_{21})$. Minimisation of Eq. (2.26) with respect to the variational parameters Q_h and Q_2 leads to the set of Euler-Lagrange equations

$$k = \frac{1 - \alpha Q_h}{1 - \beta Q_h} , \qquad (2.27)$$

$$\frac{1}{2}(\tau+\kappa^2)Q_h + 3(Q_h^2 - Q_2^2) + 4Q_h(Q_h^2 + Q_2^2) + \frac{\kappa^2}{4}Q_2^2\left[-\beta k^2 + 2\alpha k\right] = 0, \qquad (2.28)$$

$$\frac{1}{2}(\tau+\kappa^2)Q_2 - 6Q_hQ_2 + 4Q_2(Q_h^2+Q_2^2) + \frac{\kappa^2}{2}Q_2\big[(1-\beta Q_h)k^2 - 2(1-\alpha Q_h)k\big] = 0.$$
(2.29)

Using these, and assuming $Q_2 \neq 0$ as expected for a first order transition, we obtain the pair of equations

$$Q_2^2 = \frac{1}{8} \left(12Q_h + \kappa^2 \frac{(1 - \alpha Q_h)^2}{1 - \beta Q_h} - (\tau + \kappa^2) \right) - Q_h^2 , \qquad (2.30)$$

$$18Q_h^2 + \kappa^2 Q_h \frac{(1-\alpha Q_h)^2}{1-\beta Q_h} = Q_2^2 \left(6 - \frac{\kappa^2}{2} \left[2\alpha - \beta \frac{1-\alpha Q_h}{1-\beta Q_h} \right] \frac{1-\alpha Q_h}{1-\beta Q_h} \right),$$
(2.31)

which formally define the equilibrium **Q**-tensor. In general these equations must be solved numerically; however, they can be easily solved when $\alpha = \beta = 0$, and we recover the usual results [11,42,61]

$$Q_h = \frac{9-\kappa^2}{48} \left(1 + \sqrt{1 - \frac{72\tau}{(9-\kappa^2)^2}} \right), \qquad (2.32)$$

$$Q_2^2 = \frac{27+\kappa^2}{24}Q_h - \frac{3\tau}{32} . (2.33)$$

The main feature which arises from the inclusion of higher order terms in the gradient free energy is that the helical wavevector now depends on the temperature. As Eq. (2.27) shows, the wavevector depends on the amplitude of the homogeneous part of the order parameter, and since the order parameter depends on the temperature, it follows that so does the cholesteric pitch. It should be noted that, even with such a simple modification of the free energy, we can already account for the inversion of the helical sense with decreasing temperature observed experimentally in some cholesterics [1,62,63]. According to the present theory the cholesteric wavevector changes sign when $Q_h = 1/\alpha$. When this value is substituted into Eqs. (2.30) and (2.31) we obtain the following expression for the inversion temperature

$$\tau_{\rm HI} = \frac{4}{\alpha^2} (3\alpha - 8) - \kappa^2 . \tag{2.34}$$

At the inversion itself we find $Q_2 = \sqrt{3}Q_h$ and

$$\mathbf{Q} = \frac{2}{\alpha\sqrt{6}} \begin{pmatrix} -1 & 0 & 0\\ 0 & 2 & 0\\ 0 & 0 & -1 \end{pmatrix} , \qquad (2.35)$$

which corresponds, as expected, to a uniaxial nematic with director $\mathbf{n} = (0, 1, 0)$. Other orientations of the nematic within the *yz*-plane may be obtained by adding a constant phase shift to the sin and cos terms in \mathbf{Q} in Eq. (2.25). It is clear that within this theory, as observed experimentally, the helical inversion is a smooth cross-over that does not involve any discontinuities in physical quantities, i.e., it is not a phase transition.

The isotropic-cholesteric transition temperature is obtained by supplementing Eqs. (2.30) and (2.31) with the condition that the free energy (Eq. (2.26)) be zero. A short calculation leads to the following equation for Q_h at the transition temperature:

$$\left(4Q_h + \frac{\kappa^2}{4} \frac{(1-\alpha Q_h)^2}{1-\beta Q_h} \right) \left(6 - \frac{\kappa^2}{2} \frac{1-\alpha Q_h}{1-\beta Q_h} \left[2\alpha - \beta \frac{1-\alpha Q_h}{1-\beta Q_h} \right] \right)^2 - \left(24Q_h + \frac{\kappa^2 (1-\alpha Q_h)^2}{1-\beta Q_h} - \frac{\kappa^2 Q_h}{2} \frac{1-\alpha Q_h}{1-\beta Q_h} \left[2\alpha - \beta \frac{1-\alpha Q_h}{1-\beta Q_h} \right] \right)^2 = 0 .$$
 (2.36)



Figure 2.1: (a) Isotropic cholesteric transition temperature as a function of chirality for a range of parameter values: (i) $\alpha = \beta = 0$, (ii) $\alpha = 2$, $\beta = 0$, (iii) $\alpha = 2$, $\beta = 1$, (iv) $\alpha = 4$, $\beta = 0$. (b) Temperature dependence of the cholesteric wavevector for the same set of parameters as in (a) (all cases are for $\kappa = 0.05$).

Again when $\alpha = \beta = 0$ we recover the usual results [11, 42, 61]

$$Q_h^{\rm IC} = \frac{1}{8} \left(1 - \frac{\kappa^2}{3} + \sqrt{1 + \frac{\kappa^2}{3}} \right) \,, \tag{2.37}$$

$$\tau_{\rm IC} = \frac{1}{2} \left(1 - \kappa^2 + \left(1 + \frac{\kappa^2}{3} \right)^{3/2} \right) \,, \tag{2.38}$$

where $\tau_{\rm IC}$ is the temperature at which the isotropic-cholesteric transition occurs. A situation of some experimental interest occurs when the cholesteric undergoes helix inversion at the isotropic-cholesteric transition temperature [62]. This case may be solved exactly by setting $Q_h = 1/\alpha$ in Eq. (2.36). We find $\alpha = 4$ and

$$\tau_{\rm HI} = \tau_{\rm IC} = 1 - \kappa^2 \,, \tag{2.39}$$

which is precisely the transition temperature for a nematic (with non-zero κ). This particular case provides a guide to the range of values that α can be expected to take in experimental systems.

In the general case Eq. (2.36) can be solved numerically. Fig. 2.1(a) shows the isotropiccholesteric transition temperature as a function of chirality for a selection of values of α and β . The general trend can be understood on the basis of the effect that the parameters have on the helical pitch. Increasing α increases the pitch and shifts the liquid crystal towards nematic behaviour, while increasing β decreases the pitch and hence shifts away from the nematic state. The temperature dependence of the helical wavevector is shown for the same set of values of α and β in Fig. 2.1(b). It can be seen that the wavevector varies approximately linearly with temperature except in the immediate vicinity of the transition temperature. It may at first seem unusual that increasing β should lead to a larger temperature dependence, however this can be understood by noting that the inversion temperature depends only on α and that increasing β leads to a larger wavevector at the isotropic transition temperature.

Finally, it is known that if the chirality is sufficiently large then the isotropic-cholesteric transition is second order instead of first [11, 42]. We can calculate the maximum value of κ compatible with a first order transition by setting $Q_h = 0$ in Eq. (2.36). The resulting equation is solved to obtain

$$\kappa_{\max} = \frac{2}{2\alpha - \beta} \left[-1 + \sqrt{1 + 3(2\alpha - \beta)} \right].$$
(2.40)

In what follows it will be seen that this limit is never reached due to the intervention of blue phases.

2.3 Blue phases

The platelet-like visual appearance and Bragg scattering of blue phases I and II suggest that they may be well approximated by a periodic \mathbf{Q} -tensor. It is therefore convenient to introduce a Fourier decomposition [42,60]

$$\mathbf{Q}(\mathbf{r}) = \sum_{\mathbf{k}} N_k^{-1/2} \sum_{m=-2}^2 Q_m(k) e^{i\psi_m(\mathbf{k})} \mathbf{M}_m(\hat{\mathbf{k}}) e^{-i\mathbf{k}\cdot\mathbf{r}} .$$
(2.41)

Here Q_m, ψ_m are the amplitude and phase of the Fourier mode, N_k is a convenient normalising factor equal to the number of Fourier modes **k** that all have the same magnitude k, and the tensors \mathbf{M}_m represent a set of basis tensors adapted to each Fourier mode. Reality of the **Q**-tensor imposes the constraint

$$e^{-i\psi_m(\mathbf{k})}\mathbf{M}_m^{\dagger}(\hat{\mathbf{k}}) = e^{i\psi_m(-\mathbf{k})}\mathbf{M}_m(-\hat{\mathbf{k}}) , \qquad (2.42)$$

which we choose to satisfy by requiring separately that $\psi_m(-\mathbf{k}) = -\psi_m(\mathbf{k})$ and $\mathbf{M}_m^{\dagger}(\hat{\mathbf{k}}) = \mathbf{M}_m(-\hat{\mathbf{k}})$. The basis tensors $\mathbf{M}_m(\hat{\mathbf{k}})$ should themselves be chosen so as to diagonalise the quadratic part of the Landau-de Gennes free energy. This calculation was first done by Bra-

zovskii and Dmitriev [60] who showed that the appropriate choice is

$$\mathbf{M}_{\pm 2} = \frac{1}{2} (\mathbf{v} \pm i\mathbf{w}) \otimes (\mathbf{v} \pm i\mathbf{w}) ,$$

$$\mathbf{M}_{\pm 1} = \frac{1}{2} \Big[(\mathbf{v} \pm i\mathbf{w}) \otimes \hat{\mathbf{k}} + \hat{\mathbf{k}} \otimes (\mathbf{v} \pm i\mathbf{w}) \Big] ,$$

$$\mathbf{M}_{0} = \frac{1}{\sqrt{6}} (3\hat{\mathbf{k}} \otimes \hat{\mathbf{k}} - \mathbf{I}) .$$
(2.43)

Here the vectors \mathbf{v}, \mathbf{w} are two unit vectors chosen such that the trio $\{\hat{\mathbf{k}}, \mathbf{v}, \mathbf{w}\}$ form a righthanded, orthonormal set. However, this choice is not unique, in the sense that the vectors \mathbf{v}, \mathbf{w} may be freely rotated about the direction $\hat{\mathbf{k}}$ and still form a right-handed, orthonormal set. Thus there is a U(1) gauge freedom in the choice of the basis tensors. Under a gauge transformation, where \mathbf{v}, \mathbf{w} are rotated through an angle Θ in the right-handed sense, the basis tensors transform as $\mathbf{M}_m \to e^{im\Theta} \mathbf{M}_m$.

The **Q**-tensor for a periodic blue phase is obtained by specifying the set of wavevectors to appear in the Fourier decomposition and the values for the phases $\psi_m(\mathbf{k})$, given for a particular choice of gauge for the tensors $\mathbf{M}_m(\hat{\mathbf{k}})$. The choice of lattice type determines the set of wavevectors, while the phases $\psi_m(\mathbf{k})$ are determined by the particular space group of the candidate blue phase. An application of Buerger's method [64] shows that the symmetry operations of the space group lead to the relations

$$\psi_m(\mathbf{Rk}) = \psi_m(\mathbf{k}) + \mathbf{k} \cdot \mathbf{t} - (\mathbf{k} - \mathbf{Rk}) \cdot \mathbf{y} + m\Theta(\mathbf{k}, \mathbf{Rk}) \mod 2\pi , \qquad (2.44)$$

between the phases of the Fourier modes. Here \mathbf{R} is the rotational part of the symmetry operation, \mathbf{t} is the translational component along the rotation axis, \mathbf{y} is the point on the rotation axis closest to the origin, and $\Theta(\mathbf{k}, \mathbf{Rk})$ is an additional phase angle coming from the gauge transformation between $\mathbf{RM}_m(\hat{\mathbf{k}})\mathbf{R}^T$ and $\mathbf{M}_m(\mathbf{R\hat{k}})$.

Eq. (2.44) places considerable restrictions on the possible space groups that a blue phase texture can have. We illustrate this for the most immediately relevant case of cubic textures, although the results also apply to tetragonal textures, which we discuss in Chapter 3. All cubic systems possess four-fold rotational symmetry about the cubic axes: we consider the effect of a four-fold rotation about the z-axis on the wavevectors $(2\pi/L_z)l\mathbf{e}_z$. In this case $\mathbf{Rk} = \mathbf{k}$ and the gauge angle $\Theta(\mathbf{k}, \mathbf{Rk})$ is simply $\pi/2$, so that Eq. (2.44) reduces to

$$2\pi lp/4 + m\pi/2 = 0 \mod 2\pi , \qquad (2.45)$$

with p denoting the order of the screw axis. For energetic reasons the m = 2 modes are the most important ones to consider [42, 60]. Restricting our attention to this case we find that if the lattice is simple cubic, l = 1, then the four-fold axis has p = 2, while if the lattice is body-centred cubic, l = 2, then the four-fold axis has p = 1. Thus generically, simple cubic textures have 4_2 screw axes and body-centred cubic textures have 4_1 screw axes. An exception is possible where the amplitude $Q_2(k)$ vanishes, which in fact occurs for the O^5 texture [42].

We do not show explicitly the remainder of the calculation of the Fourier phases, but only record the results. For blue phase I the space group is $O^{8-}(I4_132)$ and the phases are [42, 43]

$$\psi_2(110) = \mp \pi; \quad \psi_2(\bar{1}10) = \pm \pi/2, \qquad \text{plus cyclic permutations}, \qquad (2.46)$$

where we have adopted the conventional shorthand (hkl) notation for reciprocal lattice vectors. For blue phase II the space group is $O^2(P4_232)$, for which the phases are [42, 43]

$$\psi_2(100) = 0$$
, plus cyclic permutations. (2.47)

In both cases the phases are given relative to a natural gauge in which the local basis vectors \mathbf{v}, \mathbf{w} are chosen to lie along the lattice directions, e.g., for $\hat{\mathbf{k}} = \mathbf{e}_x$ we choose $\mathbf{v} = \mathbf{e}_y$ and $\mathbf{w} = \mathbf{e}_z$. It is noteworthy that two choices are possible in the case of blue phase I: in fact these correspond to distinct blue phases, each possessing different disclination networks [11]. We shall follow the convention adopted in the literature and refer to the texture arising from choosing the upper sign as O^{8-} and that from choosing the lower sign as O^{8+} .

2.3.1 Free energy of the blue phases

The Fourier decomposition, Eq. (2.41), provides a means to calculate analytically the free energy of a blue phase texture. When this form of the **Q**-tensor is inserted into the free energy,

Eq. (2.22), we obtain [42, 43]

$$\begin{split} f &= \sum_{\mathbf{k}} N_{k}^{-1} \sum_{m} Q_{m}^{2}(k) \left\{ \frac{\tau + \kappa^{2}}{4} + \frac{\kappa^{2}}{4} \left[\left(1 + \frac{4 - m^{2}}{6} \frac{L_{22} - L_{21}}{L_{21}} \right) k^{2} - m k \right] \right\} \\ &- \sqrt{6} \sum_{\mathbf{k}_{1}, \mathbf{k}_{2}, \mathbf{k}_{3}} N_{k_{1}}^{-1/2} N_{k_{2}}^{-1/2} N_{k_{3}}^{-1/2} \sum_{m_{1}, m_{2}, m_{3}} Q_{m_{1}}(k_{1}) Q_{m_{2}}(k_{2}) Q_{m_{3}}(k_{3}) \\ &\times e^{i(\psi_{m_{1}}(\mathbf{k}_{1}) + \psi_{m_{2}}(\mathbf{k}_{2}) + \psi_{m_{3}}(\mathbf{k}_{3}))} \delta_{\mathbf{k}_{1} + \mathbf{k}_{2} + \mathbf{k}_{3}} \\ &\left\{ \left[1 + \frac{\kappa^{2}}{4} \frac{2L_{38}}{9L_{21}} \left(\mathbf{k}_{2} \cdot \mathbf{k}_{3} \right) \right] \operatorname{tr} \left(\mathbf{M}_{m_{1}}(\hat{\mathbf{k}}_{1}) \mathbf{M}_{m_{2}}(\hat{\mathbf{k}}_{2}) \mathbf{M}_{m_{3}}(\hat{\mathbf{k}}_{3}) \right) \\ &+ \frac{\kappa^{2}}{4} \frac{2L_{34}}{L_{21}} \delta_{m_{2}, m_{3}} \mathbf{k}_{2} \cdot \mathbf{M}_{m_{1}}(\hat{\mathbf{k}}_{1}) \cdot \mathbf{k}_{3} - \frac{\alpha \kappa^{2}}{4} \operatorname{tr} \left(i \mathbf{k}_{3} \times \mathbf{M}_{m_{1}}(\hat{\mathbf{k}}_{1}) \mathbf{M}_{m_{2}}(\hat{\mathbf{k}}_{2}) \mathbf{M}_{m_{3}}(\hat{\mathbf{k}}_{3}) \right) \\ &+ \sum_{\mathbf{k}_{1}, \mathbf{k}_{2}, \mathbf{k}_{3}, \mathbf{k}_{4}} N_{k_{1}}^{-1/2} N_{k_{2}}^{-1/2} N_{k_{3}}^{-1/2} N_{k_{4}}^{-1/2} \sum_{m_{1}, m_{2}, m_{3}, m_{4}} Q_{m_{1}}(k_{1}) Q_{m_{2}}(k_{2}) Q_{m_{3}}(k_{3}) Q_{m_{4}}(k_{4}) \\ &\times e^{i(\psi_{m_{1}}(\mathbf{k}_{1}) + \psi_{m_{2}}(\mathbf{k}_{2}) + \psi_{m_{3}}(\mathbf{k}_{3}) + \psi_{m_{4}}(\mathbf{k}_{4})} \delta_{\mathbf{k}_{1} + \mathbf{k}_{2} + \mathbf{k}_{3} + \mathbf{k}_{4}} \\ &\times \operatorname{tr} \left(\mathbf{M}_{m_{1}}(\hat{\mathbf{k}}_{1}) \mathbf{M}_{m_{2}}(\hat{\mathbf{k}}_{2}) \right) \operatorname{tr} \left(\mathbf{M}_{m_{3}}(\hat{\mathbf{k}}_{3}) \mathbf{M}_{m_{4}}(\hat{\mathbf{k}}_{4}) \right). \end{split}$$

$$(2.48)$$

The large number of terms contributing to the cubic and quartic invariants makes an explicit calculation of this free energy for any chosen blue phase texture a very laborious process. On the small number of occasions when this has been seriously attempted [11, 42, 43, 57, 61, 65] the Fourier sum has been truncated to a small number of wavevectors and to m = 2 modes only, in order to keep the calculation manageable. The restriction to m = 2 modes is motivated from a consideration of the quadratic free energy, which is minimised by the choice m = 2. This is often justified by considering the high chirality limit, $\kappa \to \infty$, however, the conclusion is only arrived at if the gradient free energy is restricted to quadratic order in \mathbf{Q} . Furthermore, if only m = 2 modes are retained in the Fourier expansion then the free energy of the blue phase is independent of the elastic constant L_{22} so that the quadratic gradient theory shows no sensitivity to the relative magnitude of the Frank splay, twist and bend elastic constants. Since these are observed experimentally to greatly influence the blue phase stability [66] we must conclude that $m \neq 2$ modes contribute significantly to the blue phase free energy. Nonetheless, the restriction to m = 2 modes provides an appropriate first approximation, both for analytic and numerical calculations, and we shall use it to construct Q-tensors that serve as the initial condition for numerical simulations.

2.3.2 Lattice constants and the redshift

The pitch of the cholesteric helix is determined by the strength of chiral dopant. In both the Frank director and Landau-de Gennes theories this is encoded by the constant q_0 appearing in the free energy density. Similarly, the lattice constants of blue phases I and II are also determined by the strength of the chiral dopant, q_0 , however, they need not coincide with the value of the cholesteric pitch. Experimentally it is found that the blue phase lattice constants are larger than the helical pitch, typically by 10-20%. This observation is accommodated naturally within the Landau-de Gennes theory by minimising Eq. (2.48) with respect to the magnitude of the primitive reciprocal lattice constant, $2\pi/a$. The wavevectors in a cubic system have magnitude $(2\pi/a)(h^2 + k^2 + l^2)^{1/2}$, so that the reciprocal lattice constant is given by [42]

$$\frac{2\pi}{a} = \frac{\sum_{\langle h,k,l \rangle} \sum_{m} Q_m^2(h,k,l) \ (h^2 + k^2 + l^2)^{1/2} \ (m/2)}{\sum_{\langle h,k,l \rangle} \sum_{m} Q_m^2(h,k,l) \ (h^2 + k^2 + l^2)} ,$$
(2.49)

if we restrict to the quadratic gradient theory. For the cholesteric texture, the **Q**-tensor contains a single Fourier wavevector, (100) say, corresponding to the mode m = 2, so that in dimensionless units we have $2\pi/p = 1$ (Eq. (2.27) with $\alpha, \beta = 0$). Re-instating the dimensions we recover the familiar result $p = \pi/q_0$ [1].

In the infinite chirality limit, the **Q**-tensors describing crystalline blue phases contain only the fundamental set of wavevectors and we find that $2\pi/a = (h^2 + k^2 + l^2)^{-1/2}$ so that the magnitude of these wavevectors is again $k = 2q_0$ [11,42], as in the cholesteric phase. However, for any finite value of the chirality, the presence of higher harmonics will lead to the magnitude of the fundamental wavevector taking a value lower than $2q_0$. This decrease can be quantified by a parameter called the *redshift*, which is defined through the relation $k =: 2q_0 r$ between the magnitude of the fundamental wavevector and the Landau-de Gennes chiral parameter q_0 .

2.4 Numerical minimisation: the cholesteric phase diagram

We now describe the phase diagram resulting from a minimisation of the modified Landau-de Gennes free energy, Eq. (2.22), taking into account the two cubic blue phases. Some progress in obtaining an analytic description of the blue phases is possible, see, e.g., Reference [65], however, this approach involves adopting an approximate form for the order parameter and only gives a constrained minimisation. Furthermore, although the analytic theory correctly identifies the two structures observed in experiments, it does not reproduce the correct order of appearance of the two phases at low chiralities. A full minimisation can be achieved numerically and has been described in Reference [54] for the one elastic constant approximation.

To study the different phases, cholesteric, blue phase I or blue phase II, it is necessary to implement appropriate initial conditions for the simulation. The **Q**-tensor is initialised using analytic expressions appropriate to the high chirality limit, which act to define the symmetry of the chosen phase. These are constructed following the Fourier analysis of § 2.3, truncating the Fourier series at the fundamental set of wavevectors and restricting to m = 2 modes only. For blue phase I we use [11, 42]

$$Q_{xx} \sim -\sin(ky/\sqrt{2})\cos(kx/\sqrt{2}) - \sin(kx/\sqrt{2})\cos(kz/\sqrt{2}) + 2\sin(kz/\sqrt{2})\cos(ky/\sqrt{2}) ,$$

$$Q_{xy} \sim -\sqrt{2}\sin(kx/\sqrt{2})\sin(kz/\sqrt{2}) - \sqrt{2}\cos(ky/\sqrt{2})\cos(kz/\sqrt{2}) + \sin(kx/\sqrt{2})\cos(ky/\sqrt{2}) ,$$
(2.50)

with the other components obtained by cyclic permutation. Similarly, for blue phase II the \mathbf{Q} -tensor is initialised as [11, 42]

$$Q_{xx} \sim \cos(kz) - \cos(ky) ,$$

$$Q_{xy} \sim \sin(kz) ,$$
(2.51)

with the other components again obtained by cyclic permutation. We comment briefly that these expressions are not identical with those given in the seminal work of Grebel, Hornreich and Shtrikman [42]. This is due to a minor error in that work, which the expressions quoted here correct.

Under subsequent numerical evolution according to Eq. (1.8) the system relaxes to that structure of the same symmetry which minimises the free energy. We are therefore able to obtain, for any value of the parameters, *local* minima of the free energy corresponding to each of the cholesteric and blue phases. The *global* free energy minimum was taken to be the smallest of these calculated local minima.

We have seen in § 2.2 that the inclusion of cubic invariants in the gradient free energy leads to a temperature dependent helical pitch in the cholesteric phase, Eq. (2.27). For the blue phases as well, the unit cell size is temperature dependent, so that to achieve a full minimisation of the free energy it is necessary to set the correct unit cell size in the simulation. This unit cell size is not known *a priori*, but rather depends on the magnitude of the order parameter, a quantity that is only determined by the numerical minimisation. Therefore we must introduce a means of determining, and setting, the unit cell size as the **Q**-tensor evolves during the simulation. We can account for a change in unit cell size by rescaling the gradient contributions to the free energy and molecular field. This is accomplished in practice by changing the elastic constants as follows

$$q_0 = q_0^{\text{init}} / r ,$$

$$L_{2a} = L_{2a}^{\text{init}} \times r^2 ,$$

$$L_{3b} = L_{3b}^{\text{init}} \times r^2 ,$$
(2.52)

where a = 1, 2, b = 1, ..., 9, a superscript 'init' denotes the initial value of a simulation parameter and r is the appropriate rescaling factor, which is identical to the redshift described in § 2.3.2. One shortcoming of analytic calculations [42, 43] is that the value of the redshift is not determined exactly, but only for the approximate form of the **Q**-tensor that is assumed. Similarly, in previous numerical work [54], the redshift was assumed to take the value suggested by the approximate analytic calculations. The exact redshift for the cholesteric phase could be calculated by obtaining the numerical value of Q_h and using Eq. (2.27). However, a similar approach is not available for the blue phases and consequently it is more useful, and easier, to calculate it using the free energy.

To do this we note that since the free energy is quadratic in gradients, it may be written formally in \mathbf{k} -space as

$$f = ak^2 + bk + c , (2.53)$$

where the coefficients a, b and c depend on the **Q**-tensor, but not on **k**. The optimum wavevector is given by k = -b/2a, and since the coefficients a and b are determined by the simulation at every timestep it is straightforward to use these values to determine the exact value for the redshift. This is not a dynamic process: the redshift is simply set to this optimal value and does not relax towards equilibrium.

In order to verify that the procedure was working successfully, and to check the level of accuracy that could be obtained, we used this numerical method to calculate the wavevector of a cholesteric undergoing helix inversion and compared it to the theory described in § 2.2.



Figure 2.2: Comparison of numerical (\diamond) and analytic (solid line) calculations of the helical pitch in a cholesteric displaying helical sense inversion. (a) Cholesteric wavevector and (b) pitch against reduced temperature, τ .

Simulation parameters were chosen to set $\alpha = 2.0$ and $\kappa \approx 0.096$, leading to an inversion temperature of $\tau_{\rm HI} \approx -2$, Eq. (2.34). The results are shown in Fig. 2.2. We have plotted both the helical wavevector, which is the relevant quantity theoretically, and the pitch, since this is more frequently given in experimental work. As can be seen the agreement is excellent even very close to the inversion point.

2.4.1 Phase diagram

The phase diagram for chiral liquid crystals obtained for a selection of parameter values using the extended Landau-de Gennes free energy, Eq. (2.21), is shown in Fig. 2.3. These were obtained numerically using a lattice Boltzmann algorithm [32] to solve the Beris-Edwards equations for the evolution of \mathbf{Q} , Eqs. (1.8) - (1.12). A single unit cell was simulated and periodic boundary conditions imposed in all directions. We show first, in Fig. 2.3(a), the phase diagram obtained for the one elastic constant approximation with numerical optimisation of the unit cell size. For comparison, the phase diagram for a fixed unit cell size determined in Reference [54] is reproduced in Fig. 2.3(b).

Although the qualitative features remain unchanged, we note that optimisation of the unit cell size has extended the range of stability of blue phase I, both relative to the cholesteric phase and relative to blue phase II. The movement of the cholesteric phase boundary is quite significant, with the triple point moving to lower chirality by about 20%. This is due to the optimum redshift taking a lower value at these chiralities than was assumed previously. Based on analytic calculations of Grebel *et al* [42, 43] the redshift was assigned the value 0.79 in Reference [54]. However, we find a much lower value, with average 0.68 at the cholesteric-blue phase I phase boundary (the value is roughly independent of temperature, except very close to the isotropic transition temperature). In contrast, at the blue phase I-blue phase II phase boundary we obtain an average redshift of 0.77 for blue phase I and 0.86 for blue phase II. These values are in better agreement with the analytic results of Grebel *et al* [42,43], primarily because the chirality at this phase boundary is much closer to the values of chirality where the analytic calculations predict the blue phases are stable. In what follows we will use Fig. 2.3(a) as a reference phase diagram relative to which the effect of varying the Landau-de Gennes parameters can by measured.

We now consider how the phase diagram changes as the ratios of the elastic constants are varied. Here one expects that the qualitative features of the phase diagram will be retained, but it is nonetheless of interest to determine how large a quantitative shift can be obtained. Fig. 2.3 shows the phase diagrams obtained upon separately varying the twist and bend elastic constants.

To investigate the effect of the bend elastic constant we chose parameter values $L_{21} = L_{22} =$ $L_{34} = 0.02, L_{38} = 0$ which corresponds to a ratio of bend to splay of about 1.75, while splay and twist remain degenerate. The stability of blue phase I is seen to decrease quite significantly relative to the cholesteric phase while at the same time there is a small increase in stability over blue phase II. There is only a minor shift in the cholesteric-blue phase I phase boundary at the transition temperature, however, as the temperature decreases the shift becomes larger. For example at a reduced temperature of $\tau = -2$ the phase boundary occurs at a chirality of $\kappa \approx 1$, representing a shift to higher chiralities of almost 70% as compared to the one elastic constant case. The value of the redshift is markedly increased, with the average value for blue phase I at the cholesteric boundary being 0.81. However, it was shown in \S 2.2 that the cholesteric wavevector depends on the value of L_{34} and hence has also increased. This raw value of the redshift no longer represents the most directly accessible quantity and a more relevant figure is the ratio of the blue phase I redshift to that of the cholesteric, since this can be measured experimentally by means of the discontinuity of the back-scattered Bragg peak. For this quantity we obtain an average of 0.67 and note the strong similarity of this value with that obtained in the one elastic constant approximation.

At the blue phase I-blue phase II phase boundary the redshift is 0.87 and 0.97 for blue phase I and blue phase II respectively. Again, it is the ratio of these values which is more directly



Figure 2.3: Numerical evaluation of the phase diagram of cholesteric liquid crystals within the Landau-de Gennes theory. τ , the reduced temperature, and κ , the chirality, are defined by Eqs. (2.23) and (2.24). Phase diagrams in the one elastic constant approximation with (a) optimisation of the unit cell size, and (b) fixed unit cell size, after [54]. Phase diagrams with unequal elastic constants: (c) $K_1 = K_2 = 0.5K_3$, (d) $K_1 = K_3 = 1.5K_2$. (e) The numerical phase diagram obtained with the chiral invariant, Eq. (2.8), added to the free energy. The magnitude of this term was chosen so as to produce helical sense inversion in the cholesteric phase at a temperature not far below the isotropic transition temperature. (f) An enlargement of the region near the isotropic transition temperature. Note the reversal in the order of appearance of blue phase I and blue phase II as a function of chirality.
relevant to experiment. The ratio of the O^{8-} unit cell size to that of O^2 is 0.90, and again we note a strong similarity with the ratio obtained from the one elastic constant approximation, 0.89.

The value of the twist elastic constant is controlled by the Landau-de Gennes parameter L_{22} . In most liquid crystals the twist elastic constant is smaller than either splay or bend. In order to match this, we constructed the phase diagram for parameter values $L_{21} = 0.02$, $L_{22} = 0.04$, $L_{34} = L_{38} = 0$, which is shown in Fig. 2.3(d). This choice of parameters resulted in a ratio of splay to twist of about 1.5, while splay and bend remained degenerate. Again we observe that the stability of blue phase I is reduced relative to the cholesteric phase by an amount similar to that seen by varying the bend elastic constant. The values of the redshift for both blue phase I and blue phase II are only very slightly increased relative to their values in the one elastic constant limit, while the cholesteric wavevector is insensitive to the value of L_{22} . This reveals an intriguing feature, that while the phase boundaries and absolute values of the redshift can vary appreciably, the ratios of the redshift for the different phases, and hence the discontinuities in back-scattered Bragg peaks, are essentially independent of the values of the values of the elastic constants.

Finally, we consider the effect of the chiral cubic invariant on the blue phases. We chose parameter values of $L_{21} = L_{22} = L_{34} = 0.02$, $L_{38} = 0.04$, $\alpha = 2.0$, which for the cholesteric phase sets $\beta = 0$ and gives a ratio of bend to splay of about 1.6. The phase diagram is shown in Fig. 2.3(e).

The value of α is such that the cholesteric undergoes helical sense inversion at a reduced temperature of about $\tau \approx -2$. What is remarkable is the dramatic increase in stability of blue phase I relative to the cholesteric phase. The region of stability has been increased down to chiralities as low as $\kappa = 0.07$ and at such low chiralities the phase boundary is essentially independent of κ for all τ . In addition, we find a very small region of stability for blue phase II located close to the isotropic transition. As shown in greater detail in Fig. 2.3(f) this occurs in a narrow temperature interval at chiralities lower than those for which blue phase I is stable, representing a reversal of the order of appearance of the two blue phases. In the region where blue phase II is stable we find a redshift of 0.36. In contrast to the situation without the chiral invariant, this is *smaller* than the blue phase I redshift, which takes the value 0.43. Again, the absolute values of the blue phase redshifts are not as relevant as their ratios to that of the cholesteric phase. In this case we find the cholesteric redshift is 0.49 at the isotropic transition



Figure 2.4: Numerical results for the temperature dependence of the (a) wavevector and (b) pitch of the unit cell of blue phase I along the cholesteric-blue phase I phase boundary at $\kappa \approx 0.07$ (\diamond). For comparison the cholesteric wavevector and pitch are also given: numerical results (\circ), results from the calculation presented in § 2.2 (solid line).

temperature, giving ratios of 0.88 for blue phase I and 0.74 for blue phase II, both of which are significantly different to those obtained in the one elastic constant limit.

Since blue phase I is now stable over a much larger temperature range it displays a significant variation in unit cell size as the temperature is lowered. As an illustration of this, the blue phase I redshift is 0.19 at a reduced temperature of $\tau \approx -5$, corresponding to more than a two-fold increase in the lattice parameter. A plot of the temperature dependence of the blue phase I redshift is shown in Fig. 2.4. For comparison the helical wavevector of the cholesteric phase is also plotted on the same graph.

2.4.2 Discussion

We have investigated numerically the phase diagram of the cholesteric blue phases for a range of parameter values within the framework of an extended Landau-de Gennes theory. The traditional Landau-de Gennes theory has long been known to only accommodate two independent Frank elastic constants and to have a temperature independent cholesteric pitch. Both of these shortcomings were overcome by retaining terms of cubic order in the **Q**-tensor in the expansion for the gradient free energy. Since the new terms were added specifically to remove the degeneracy between splay and bend, and to give a temperature dependence to the helical pitch they possess a clear and simple physical interpretation. In particular, the value of the parameter α , which controls the strength of the chiral cubic invariant, should be relatively easy to estimate on the basis of Eq. (2.34) for the helix inversion temperature. The magnitude of the elastic constants for the achiral cubic invariants is more difficult to ascertain. Although this may be estimated from the ratios of the Frank elastic constants it is clear that, because there are more Landau-de Gennes elastic constants than Frank, the latter are insufficient to uniquely determine the former (an estimation of the magnitude of the cubic Landau-de Gennes elastic constants was given in Reference [56]).

The extended Landau-de Gennes theory that we have investigated provides a phenomenological description of helical sense inversion in the cholesteric phase. The inversion arises as a natural consequence of the presence of including higher order chiral invariants in the gradient free energy. Although such higher order contributions are usually neglected since they are deemed small compared to the terms already retained, the fact that helix inversion is observed experimentally demonstrates that these terms can play a significant role. We also note that, for systems undergoing first order phase transitions, such as liquid crystals, although the order parameter is small it is not infinitesimal, and therefore the relative magnitude of a given term in the Landau expansion depends not only on the power of the order parameter but also on the size of any numerical coefficient premultiplying it.

The primary aim of this Chapter has been to investigate how much the properties of the blue phases can be changed within the framework of Landau-de Gennes theory. In this regard we have shown that the retention of cubic order terms in the gradient free energy can lead to considerable changes in the size of the blue phase unit cell and in their phase diagram. Most dramatic amongst the results is the increase in stability of blue phase I obtained in systems where the cholesteric undergoes helical sense inversion. Again we comment that this significant, qualitative change in the phase diagram arises from retaining cubic order terms and demonstrates that these give rise to more than just small changes in the physical properties.

It is of interest to consider whether the mechanism considered here is a candidate to account for the increased range of stability in blue phase I recently reported by Coles and Pivnenko [50]. It seems not, as apart from the large temperature range most features of their blue phase differ from those obtained for the choice of parameters we made here. For example, the numerics show an increase in the size of the blue phase I unit cell with decreasing temperature, while in the experimental system the unit cell size shows a small decrease. Also, numerically we find that blue phase II has a larger unit cell than blue phase I at the transition between the two, in contrast to what is found experimentally. However, we remark that the parameter space in the Landau-de Gennes theory is large enough that these discrepancies could well be resolved by a different choice of parameters.

2.5 Real space structure of the blue phases

We conclude this Chapter with a brief description of the real space structure and texture of the cubic blue phases, partly for completeness, partly to emphasise some features of double twist cylinders not mentioned in the extant literature and partly to explain the meaning of the figures we will show in Chapters 3 and 4. We focus on blue phase II for simplicity, but also provide numerical results for blue phase I.

The approximate expressions for the **Q**-tensor, Eqs. (2.50) and (2.51), contain only the fundamental set of Fourier wavevectors and are restricted to the m = 2 modes. As such, they do not lead to accurate estimates of the free energies of the cubic blue phases. In particular, at this level of truncation, the expression for blue phase II, Eq. (2.51), yields a vanishing cubic invariant and thus predicts a *second* order transition from the isotropic phase. Nonetheless, because they capture the correct symmetries, the simple expressions given in Eqs. (2.50) and (2.51) provide a remarkably accurate description of the texture of the cubic blue phases. In particular, they correctly give the location and structure of both the disclination network and the double twist cylinders; the two most identifying features of the blue phases. We outline this for the case of blue phase II.

2.5.1 Disclination lines

The location of the disclinations may be identified from the requirement that the maximal eigenvalue of \mathbf{Q} is degenerate so that the director field is undefined. Eq. (2.2) leads to the condition

$$\left(3 - \cos(kx)\cos(ky) - \cos(ky)\cos(kz) - \cos(kz)\cos(kx)\right)^3 = 27\sin^2(kx)\sin^2(ky)\sin^2(kz),$$
(2.54)

which can be seen by inspection to be satisfied everywhere along the body diagonals, $x, y, z = \pm (2\pi/k)t$. Along the [111] body diagonal the **Q**-tensor takes the form

$$\mathbf{Q} \sim \sin(2\pi t) \begin{pmatrix} 0 & 1 & 1 \\ 1 & 0 & 1 \\ 1 & 1 & 0 \end{pmatrix} , \qquad (2.55)$$

so that in a frame in which \mathbf{Q} is diagonal we have

$$\mathbf{Q} \sim \sin(2\pi t) \operatorname{diag}(2, -1, -1)$$
. (2.56)

From this we see that the subleading eigenvalues are degenerate for 0 < t < 1/2 and the **Q**-tensor is uniaxial, but there is no defect, while for 1/2 < t < 1 the maximal eigenvalue is degenerate indicating the presence of a disclination line. The network of these disclination lines extending along half of the body diagonals is shown in Fig. 2.5.

It is easier to identify the disclinations numerically from the magnitude of the order (more correctly, of the maximal eigenvalue of \mathbf{Q}) than from the criterion that two eigenvalues are degenerate. Within the defect cores, the large energy cost of the disclination is relieved by reducing the magnitude of the order, allowing the core region to become partially isotropic. Thus in all our images of the disclination network the defects are identified by plotting an isosurface, everywhere within which the magnitude of the order drops below a threshold value.

2.5.2 Double twist cylinders

Unlike the disclination lines, there is no mathematical procedure analogous to Eq. (2.2) to identify double twist cylinders in a blue phase texture and our only guides are symmetry and past experience. Here, we propose to identify the axes of the double twist cylinders using the criterion that the director is parallel to one of the cubic directions everywhere along the cylinder axis. We shall sketch in outline that this correctly identifies the double twist structure of blue phase II, and show in Chapter 4 that this also extends to the deformation of the cylinders under an applied electric field.

We consider in turn the three lines (1/2, 0, t), (t, 0, 1/2) and (1/2, t, 1/2). For the first line we have

$$\mathbf{Q}(1/2,0,t) \sim \begin{pmatrix} -1 + \cos(2\pi t) & \sin(2\pi t) & 0\\ \sin(2\pi t) & -1 + \cos(2\pi t) & 0\\ 0 & 0 & 2 \end{pmatrix} .$$
(2.57)

The eigenvalues are $0, \pm 2$, so that **Q** is maximally biaxial, and the director is given by $\mathbf{n} =$

(0,0,1) $\forall t$. Along the second line we find

$$\mathbf{Q}(t,0,1/2) \sim \begin{pmatrix} -2 & 0 & 0\\ 0 & 1 + \cos(2\pi t) & \sin(2\pi t)\\ 0 & \sin(2\pi t) & 1 - \cos(2\pi t) \end{pmatrix}, \qquad (2.58)$$

so that again the eigenvalues are $0, \pm 2$, but this time the director is not constant and instead takes the form $\mathbf{n} = (0, \cos(\pi t), \sin(\pi t))$. Finally, along the third line the **Q**-tensor is given by

$$\mathbf{Q}(1/2, t, 1/2) \sim \begin{pmatrix} -1 - \cos(2\pi t) & 0 & \sin(2\pi t) \\ 0 & 0 & 0 \\ \sin(2\pi t) & 0 & 1 + \cos(2\pi t) \end{pmatrix} .$$
(2.59)

The eigenvalues are now $0, \pm 2\cos(\pi t)$ and the director, in the region about t = 0, is given by $\mathbf{n} = (\sin(\pi t/2), 0, \cos(\pi t/2)).$

The structure we have just described is, of course, a double twist cylinder with the line (1/2, 0, t) describing the cylinder axis. However, unlike the double twist cylinders which form the basis of the low chirality theories of blue phases, this cylinder is biaxial and not uniaxial. Consequently its cross-section is elliptical instead of circular. A numerical visualisation of the double twist cylinders throughout the unit cell, shown in Fig. 2.5, reveals further that this cross-section is not uniform along its axis, but shows a periodic modulation commensurate with the lattice. Here we are showing an isosurface everywhere inside which the director has a projection onto one of the cubic directions that is larger than some threshold value. Finally, we comment that in the case of blue phase I the cylinder axes are not straight lines, as has been commonly assumed in the literature.



Figure 2.5: Network of disclinations and arrangement of double twist cylinders in blue phase I (a) and blue phase II (b). The disclinations are shown on the left in blue and the double twist cylinders on the right in grey. In all figures 2^3 unit cells have been shown to more clearly illustrate the structure.

CHAPTER 3

Cubic blue phases in electric fields

When an external electric field is applied to a liquid crystal it provides a preferred spatial direction to which the director field can couple, allowing the orientation of the liquid crystal to be controlled. In a dielectric medium an electric field contributes an amount $(-\epsilon_0/2)E_{\alpha}\epsilon_{\alpha\beta}E_{\beta}$ to the free energy density. For a uniaxial material, such as a nematic liquid crystal, the dielectric tensor has different magnitudes parallel and perpendicular to the director and may be written

$$\epsilon_{\alpha\beta} = \epsilon_{\parallel} n_{\alpha} n_{\beta} + \epsilon_{\perp} \left(\delta_{\alpha\beta} - n_{\alpha} n_{\beta} \right) \,. \tag{3.1}$$

Thus in a uniaxial nematic an applied electric field couples to the director via a term in the free energy density of the form

$$\mathcal{F}_{\text{dielectric}} = \frac{-\epsilon_0(\epsilon_{\parallel} - \epsilon_{\perp})}{2} \left(\mathbf{E} \cdot \mathbf{n} \right)^2.$$
(3.2)

If the dielectric anisotropy is positive ($\epsilon_{\parallel} > \epsilon_{\perp}$) then the director will attempt to re-align parallel to the field, whilst if it is negative the director will prefer to be everywhere perpendicular to **E**.

Since the cholesteric blue phases have an orientational order which displays cubic symmetry with a lattice constant of several hundred nm they have the potential to serve as lasers, fast light modulators or photonic crystals [46–49]. Moreover, because they are fluids, their structure can be easily manipulated and hence the properties of blue phase devices are widely tunable. A natural way of seeking to do this is through the application of an external electric field. However, because of the intricacy of their director field configurations, the response of the blue phases to an applied electric field is more complicated than that of a uniaxial nematic.

Experimental work in the 1980s and 1990s identified the principal features to be electrostriction, a continuous distortion of the shape and size of the unit cell, and a series of field induced textural transitions [53,67–75]. The electrostriction involves a shift of the back-scattered Bragg peak of 5 - 10%, and is quadratic in the field strength. The direction of the shift is observed to change with the sign of the dielectric anisotropy, but blue phase I also displays an unusual response referred to as *anomalous electrostriction*, where an expansion is seen when the field is oriented parallel to the [011] direction, but a contraction for fields parallel to [001].

At larger field strengths new blue phases appear, possessing tetragonal or hexagonal symmetry [53,70–75]. The possibility of a transition to a two-dimensional hexagonal texture was first proposed theoretically by Hornreich, Kugler and Shtrikman [76], shortly after which field induced transitions were reported experimentally [70]. Three distinct field induced textures were identified, possessing tetragonal, screw hexagonal and two-dimensional hexagonal symmetry with increasing field strength [53,70–72].

Much of the electric field behaviour has been understood via extensions of the Landau-de Gennes theory [76–81], providing qualitative agreement for the quadratic field dependence of the electrostriction and the sensitivity to the sign of the dielectric anisotropy. The qualitative features of the field induced hexagonal phases, including both the screw and two-dimensional textures, have also been reproduced [79]. However, the approximations necessary in performing semi-analytic calculations lead to predictions for the electrostriction coefficients which are an order of magnitude too small, limiting the quantitative comparison which is possible. At a qualitative level, the anomalous electrostriction of blue phase I was not found within the Landau-de Gennes theory [78,80,81] and the tetragonal texture known as blue phase X has not been included in any previous theoretical calculations [79]. Moreover, all previous studies have been restricted to statics and it has not been possible to investigate changes in the director field nor the pathway by which disclinations rearrange during textural transitions.

In this Chapter we describe a numerical investigation of the effects of an electric field on the cubic blue phases by using a three-dimensional lattice Boltzmann algorithm to solve the hydrodynamic equations of motion. We present results for the electrostriction in good agreement with experiment, obtaining, in particular, the correct order of magnitude for the shift in Bragg peak and the anomalous electrostriction of blue phase I. Furthermore, we are able to follow the rearrangement of defects and unwinding of the cubic blue phases at large field strengths, and, by reproducing the relevant experimental set-up, we can investigate for the first time the continuous blue phase I-blue phase X transition.

3.1 Electrostriction

The electrostriction, or change in size and shape, of the blue phase unit cell may be described by a minimal extension of the Landau-de Gennes theory, first introduced by Lubin and Hornreich [77]. A dielectric term

$$\mathcal{F}_{\text{dielectric}} = -\Delta \epsilon \ E_{\alpha} Q_{\alpha\beta} E_{\beta} \ , \tag{3.3}$$

is added to the usual free energy density and the equilibrium state obtained by minimisation. In doing this, the high chirality approximation of the \mathbf{Q} -tensor is generalised to describe the non-cubic geometries induced by the symmetry breaking of the electric field. For small field strengths the distorted unit cell geometry may be described by relating the primitive lattice vectors under an applied electric field to those in zero field according to

$$\mathbf{a}_{i}(E) = \left(\delta_{ij} + R_{ij\alpha\beta}E_{\alpha}E_{\beta} + o(E^{4})\right)\mathbf{a}_{j}(0), \qquad (3.4)$$

where $\{\mathbf{a}_i\}$ are the set of primitive lattice vectors and $R_{ij\alpha\beta}$ are the components of the electrostriction tensor. The absence of a term linear in E_{α} in Eq. (3.4) can be argued both because typical experiments are performed using an ac field, for which the time averaged linear term vanishes, and because there are no non-zero third rank tensors $P_{ij\alpha}$ compatible with the point symmetry (432) possessed by the cubic blue phases [69].

The approximate, high chirality form of the **Q**-tensor is taken to be the same as in the absence of an electric field, but constructed with respect to the distorted lattice and with a homogeneous component Q_h added so that the dielectric term yields a non-zero contribution to the free energy. The values of the components of the electrostriction tensor are then determined by treating them as variational parameters in the Landau-de Gennes free energy. Previous theory [77, 78, 81] has shown that for the cubic blue phases with point symmetry (432) there are only three independent components of the electrostriction tensor, which we take to be $R_1 \equiv R_{zzzz}, R_2 \equiv R_{yyzz}$ and $R_3 \equiv 2R_{yzyz}$. When the field is applied parallel to the [001]

direction the distorted lattice vectors are given by

$$\mathbf{a}_{x,y}(E) = (1 + R_2 E^2) \mathbf{a}_{x,y}(0) , \qquad (3.5)$$

$$\mathbf{a}_{z}(E) = (1 + R_{1} E^{2}) \mathbf{a}_{z}(0) , \qquad (3.6)$$

so that this configuration allows for two of the components of the electrostriction tensor to be determined. The final component, R_3 , can be obtained by applying the field parallel to the [011] direction, for which the lattice vectors become

$$\mathbf{a}_{x}(E) = (1 + R_2 E^2) \mathbf{a}_{x}(0) , \qquad (3.7)$$

$$\mathbf{a}_{y,z}(E) = \left(1 + \frac{1}{2} (R_1 + R_2) E^2 \right) \mathbf{a}_{y,z}(0) + \frac{1}{2} R_3 E^2 \mathbf{a}_{z,y}(0) .$$
(3.8)

The anomalous electrostriction of blue phase I corresponds to the condition $R_1/R_3 < 0$. Experimentally it has been found that, for positive dielectric anisotropy, $R_3 > 0$ for both blue phases, $R_1 < 0$ and $R_2 > 0$ for blue phase I, while $R_1 > 0$ and $R_2 < 0$ for blue phase II [53]. Typical magnitudes of the components of the electrostriction tensor are $\sim 1 - 3 \times 10^{-2} \mu m^2 V^{-2}$, with R_1 and R_3 approximately equal in magnitude and R_2 between 2 and 3 times smaller [53,69].

Analytic calculations using the Landau-de Gennes approach [77–81] have obtained the correct signs for the components of the electrostriction tensor in blue phase II, but not in blue phase I, so that, in particular, the anomalous electrostriction was not reproduced. The magnitudes of R_1, R_2 and R_3 are found to be an order of magnitude smaller than those observed experimentally, although the ratios are in good general agreement.

The principal reason for this discrepancy and the difficulty in performing accurate analytic calculations is that the electric field couples directly only to the homogeneous component of \mathbf{Q} . Its influence on the blue phase structure is thus indirect, occuring through the coupling between different order parameter components introduced by the cubic and quartic invariants of the bulk free energy. Thus to accurately capture the effects of the electric field it is necessary to accurately calculate the cubic and quartic invariants, which are precisely the terms that are most difficult to determine analytically. A key advantage of the numerical approach we adopt here is that it allows for an exact treatment of the effect of these invariants. In previous numerical work on blue phases in zero field this was found to significantly improve the agreement between theory and experiment [54, 82], as discussed in Chapter 2.



Figure 3.1: Variation of the free energy with varying unit cell shape, blue phase I (left) and blue phase II (right): The free energy is determined for several values of the ratio L_z/L_x parameterising the distortion (markers) and fitted to a quadratic (solid lines). In the left panel, the left and right axes correspond to E = 0.0008 and E = 0.0016 respectively. In the right panel, the left and right axes correspond to E = 0.0016 and E = 0.0031 respectively.

3.1.1 Numerical simulation

The response of the cubic blue phases to the applied electric field was determined using a lattice Boltzmann algorithm [32,34] to solve the Beris-Edwards equations for the combined evolution of the **Q**-tensor and the fluid velocity [28], Eq. (1.8) - (1.12). Hydrodynamic effects have previously been shown to have a significant effect on the dynamics of defects [18,20,21,23] and on the rheological properties of blue phases [83]. The correct incorporation of hydrodynamics may therefore be important in determining the switching of blue phases in response to an applied field, although this is not the focus of our attention in this Chapter.

The cubic blue phases were initialised using the approximate, high chirality expressions for the **Q**-tensor, Eqs. (2.50) and (2.51), suitably modified to account for the non-cubic geometry induced by the electrostriction. A single unit cell was simulated and periodic boundary conditions imposed in all directions. Values of the simulation parameters were chosen to ensure that we are far from the blue phase-cholesteric phase boundary at zero field. The value of the chirality was set to $\kappa = 0.7$ and $\kappa = 1.86$ for blue phase I and blue phase II respectively, while $\tau = 0$ throughout. This models the higher chirality of blue phase II which arises in experiments upon increasing the amount of chiral dopant. In the following time and electric field are referred to in simulation units. These can be related to physical units by using the conversion that one simulation unit corresponds to 0.01 and 0.0015 μs for blue phase I and blue phase I respectively, if the rotational viscosity is 1 P. The strength of the electric field can be inferred from the critical field required to unwind a cholesteric texture, and one simulation unit then corresponds to $\sim 350 \text{ V} \ \mu\text{m}^{-1}$. We restrict our attention to positive dielectric anisotropy, $\Delta \epsilon > 0$.

A fundamental difficulty in simulating electrostriction is in correctly accounting for the distortion of the unit cell. Within the Landau-de Gennes theory the distortion is obtained by the condition that the size and shape of the unit cell are chosen so as to minimise the free energy, F. We account for this distortion in two steps: first the shape of the simulation unit cell is fixed and we determine the value of the redshift which minimises the free energy for this fixed shape of unit cell, using the technique described in § 2.4. We then perform a series of such simulations varying the shape of the unit cell, e.g., from cubic to tetragonal. Fig. 3.1 shows exemplary results of this procedure for fields applied parallel to the [001] direction of the blue phase unit cell. The free energy is determined for several values of the ratio L_z/L_x parameterising the cubic-tetragonal distortion and fitted to a quadratic. The actual distortion at any free energy is then given by the minimum of this fit. The position of the minimum at values of L_z/L_x smaller than 1 for blue phase I and larger than 1 for blue phase II is clear evidence of anomalous electrostriction.

We first consider the case of a uniform field parallel to the [001] direction, with the field strength varied from zero up to values large enough to induce an unwinding of the blue phase structure to a uniformly aligned nematic. In order to gain a more complete picture, we have looked at both the case in which the blue phase unit cell is forced to retain its cubic symmetry and also when it is allowed to become distorted to tetragonal symmetry, accounted for by simply varying the size of the unit cell along the ([001]) z-direction. The field dependence of the blue phase lattice parameters is shown in Fig. 3.2. The response is markedly different depending on whether or not the shape of the unit cell is allowed to change. When cubic symmetry is enforced there is very little change in the unit cell size until the field strength becomes large. Furthermore, the field dependence of this shift is not quadratic even at low field strengths, although it fits quite well to a quartic. This suppression of electrostriction is a consequence of the incompatibility of a homogeneous component in an order parameter possessing cubic symmetry [78]. An expansion is seen for both blue phases, hence $R_1 > 0$ for both blue phases and the anomalous electrostriction of blue phase I is not reproduced.

When the shape of the unit cell is varied, the magnitude and qualitative features of the response change considerably. In both blue phase I and blue phase II we observe the correct



Figure 3.2: Field induced distortion of the unit cell of blue phase I (left) and blue phase II (right): In the top row the shape of the unit cell has been fixed with its initial cubic symmetry, while in the bottom row we have allowed for a distortion of the shape to give tetragonal symmetry. The lattice parameters parallel (\circ , solid line) and perpendicular (\Box , dashed line) to the field are shown relative to their zero field value.

direction of the shift in lattice parameters, i.e., the correct signs of R_1 and R_2 , [53]. The different behaviour, a contraction for blue phase I and an expansion for blue phase II, parallel to the field, is a clear indication of the anomalous electrostriction of blue phase I. This is confirmed by simulations with blue phase I and the field parallel to the [011] direction, predicting an *expansion* of the unit cell along the field direction. Remarkably, anomalous electrostriction has never been explained within the free energy in Eq. (2.21): our simulations suggest that this is due to truncation in the Fourier decomposition previously used to calculate electric field induced deformations. We also observe that the field dependence of the shift is only quadratic at low field strengths. Such behaviour has not been reported experimentally, however, it should be noted that our simulations consider only perfect, free floating single crystals and that the discrepancy may be due to the difficulty of achieving such idealised conditions experimentally.

By fitting our results for the field dependence of the lattice parameters to a quadratic function at low field strengths we can extract R_1 and R_2 . In simulation units $R_1 = -18.8 \times 10^3$, $R_2 = 9.4 \times 10^3$ for blue phase I and $R_1 = 2.7 \times 10^3$, $R_2 = -1.0 \times 10^3$ for blue phase II. Mapping these to real units we obtain values in the range $10^{-2} - 10^{-1} \mu m^2 V^{-2}$ for both blue phases, in good agreement with experiments [53]. If the distortion of the unit cell is volume preserving, R_1/R_2 is exactly -2 [78]. We find this to be the case for blue phase I, but not for blue phase II where instead we obtain $R_1/R_2 \approx -2.7$, in close agreement with the value (-2.6) experimentally reported in Reference [67]. In order to estimate R_3 we also need to consider the case when the field is applied parallel to the [011] direction and the unit cell undergoes an orthorhombic distortion. Focusing on simulations at small E we estimate $R_3 = 19.6 \times 10^3$ in simulation units for blue phase I.

3.2 Field induced unwinding

In the intermediate to large field limit, when the distortion of the size and shape of the unit cell becomes considerable, it is to be expected that this will be accompanied by a distortion of the director field and disclination network of the blue phase. As we have already commented, this is an important effect with particular relevance to field induced phase transitions and the switching dynamics of potential blue phase devices. Here we look at two scenarios: in the first we consider the disclination dynamics of blue phase II when switched to the nematic phase by a field applied parallel to the [001] direction. Then we look at the set-up which gives rise experimentally to a continuous blue phase I-blue phase X transition [72].

A large part of the qualitative behaviour may be understood on the basis of simple topological considerations. The defect networks in the cubic blue phases are composed of topologically protected s = -1/2 disclination lines. As such, they can only be unwound if two such disclinations merge into a single s = -1 disclination, since the latter is topologically unstable. As we shall see, in order to do this, the defects develop a twist, converting themselves from straight lines into helices.

The switching of blue phase II is shown in Fig. 3.3(I). In these simulations the shape of the unit cell is fixed as cubic at all times, however the dynamics are unchanged when a (fixed shape) tetragonal unit cell is used instead. The switching starts with an 'unbinding' of the bicontinuous network to give a layered arrangement of isolated s = -1/2 disclinations, oriented perpendicular to the field. These isolated disclinations then develop a twist which enables them to 'reconnect' into a new bi-continuous network. Again this 'unbinds', this time leaving an array of pairs of s = -1/2 disclinations oriented parallel to the field and wrapping around each other in a double helix configuration. Finally, these merge into an unstable s = -1 disclination which unwinds to leave a defect free nematic texture.

3.3 Textural transitions: blue phase X

Next, we consider the situation in which blue phase I is placed under an intermediate field applied parallel to the [011] direction. This is the set-up corresponding to the blue phase Iblue phase X transition observed experimentally in the 1980s [71, 72], and never understood theoretically. The electrostriction distorts the shape of the unit cell until it becomes tetragonal. Starting from this geometry, and an unperturbed blue phase I, we numerically follow the evolution of the disclination network, as shown in Fig. 3.3(II). Initially, the disclinations in the network twist, they then merge to form a transiently branched structure, which finally reorganises into a new defect network, not stable at zero field. This is a candidate structure for blue phase X, as it is a new network, found via a continuous reorganisation starting from blue phase I, and only stable in a field. Our results also predict that in our candidate blue phase X (i) the disclinations perpendicular to the field are largely unaffected, (ii) the network conforms to the space group D_4^{10} identified in Reference [72], and (iii) the double twist cylinders deform but do not break during the transition (contrary to the speculations in Reference [72]; this



Figure 3.3: Switching dynamics of (I) the blue phase II-nematic transition induced at E = 0.0094 with the field parallel to the [001] direction (vertical in the figure), and (II) the blue phase I-blue phase X transition with E = 0.006, parallel to the [011] direction (vertical in the figure). The grey tubes represent regions of the fluid inside which the magnitude of the order falls below a threshold value and thus show the network of disclinations. In (I) we have shown 2^3 unit cells for clarity, while only a single unit cell is shown in (II). From top to bottom, times (in simulation units, measured from the application of an electric field) corresponding to the snapshots are: 0 (A), 7000 (B), 45000 (C) for blue phase II and 0 (A), 80000 (B), 200000 (C) for blue phase I. Axis labels are given in fractions of the unit cell size (L_x , L_y and L_z along the x, y and z directions).



Figure 3.4: Top: director field in blue phase II for fields parallel to [001], shown in the yz-plane at 1/2 lattice parameter distance along the x-axis. (a) E = 0, (b) E = 0.008. Bottom: director field in blue phase X (E = 0.006) viewed along the tetragonal axis, showing (c) hexagonal array of double twist cylinders, and (d) stacking faults. The slices in (c) and (d) are separated by 1/4 lattice parameter distance along the z axis. Axis labels are given in fractions of the unit cell size (L_x , L_y and L_z along the x, y and z directions).

observation may be verified by experiments along the lines of those in References [84,85]).

Finally, it is also of interest to look at the static changes that occur within the unit cell. Some examples of the distortion induced in the director field are shown in Fig. 3.4. In blue phase II we see a change in the director due to unbinding of the network even for moderate values of the electric field (Fig. 3.4(a) and (b)). In the gap that opens up between the separated defects the director is well-aligned parallel to the field, representing the appearance, and growth, of nematic layers within the blue phase structure. Fig. 3.4(c) and (d) show details of the configuration of the director field in the blue phase X structure, in two planes perpendicular to the [001] direction (defects highlighted). We find that the director adopts a 'screw hexagonal' texture where layers of perfect hexagons are separated by layers of 'stacking faults'. Qualitatively, this may also explain why larger fields witness the transition to hexagonal blue phases.

3.3.1 Direct simulation of blue phase X

As an additional confirmation that the field induced blue phase I transition we have obtained numerically is indeed the transition to blue phase X observed experimentally, we would like to directly initialise the blue phase X texture and confirm that it has the same disclination network and configuration of double twist cylinders. In order to do this we need to obtain the appropriate high chirality form of the **Q**-tensor with which to initialise the simulation, which can be done by following the general procedure described in § 2.3.

Definitive experiments in the 1980s by Pieranski and Cladis [72] showed that blue phase X is centred tetragonal and can be described by the space group $D_4^{10}(I4_122)$. In order to generate the required body centred tetragonal symmetry, we take the fundamental set of wavevectors to be

$$\{\mathbf{k}\} = \left\{ \pm \frac{2\pi}{L_x} \left(\mathbf{e}_x + \mathbf{e}_y \right), \pm \frac{2\pi}{L_x} \left(-\mathbf{e}_x + \mathbf{e}_y \right), \pm \frac{2\pi}{L_x} \left(\mathbf{e}_x + \frac{L_x}{L_z} \mathbf{e}_z \right), \\ \pm \frac{2\pi}{L_x} \left(\mathbf{e}_y + \frac{L_x}{L_z} \mathbf{e}_z \right), \pm \frac{2\pi}{L_x} \left(-\mathbf{e}_x + \frac{L_x}{L_z} \mathbf{e}_z \right), \pm \frac{2\pi}{L_x} \left(-\mathbf{e}_y + \frac{L_x}{L_z} \mathbf{e}_z \right) \right\},$$
(3.9)

where L_x, L_z are the lattice constants perpendicular and parallel to the tetragonal axis, respectively. For the specification of the the basis tensors $\mathbf{M}_m(\hat{\mathbf{k}})$ we need to assign a choice of gauge to each of these wavevectors, which we do as follows

$$\mathbf{k} = (110) : \quad \mathbf{v} = \frac{1}{\sqrt{2}} \left(-\mathbf{e}_x + \mathbf{e}_y \right), \quad \mathbf{w} = \mathbf{e}_z , \qquad (3.10)$$

$$\mathbf{k} = (\bar{1}10) : \quad \mathbf{v} = \frac{1}{\sqrt{2}} \left(-\mathbf{e}_x - \mathbf{e}_y \right), \quad \mathbf{w} = \mathbf{e}_z , \qquad (3.11)$$

$$\mathbf{k} = (101)$$
 : $\mathbf{v} = \mathbf{e}_y$, $\mathbf{w} = \frac{1}{\sqrt{1 + (L_x/L_z)^2}} \left(-(L_x/L_z)\mathbf{e}_x + \mathbf{e}_z \right)$, (3.12)

$$\mathbf{k} = (011) : \mathbf{v} = -\mathbf{e}_x , \quad \mathbf{w} = \frac{1}{\sqrt{1 + (L_x/L_z)^2}} \left(-(L_x/L_z)\mathbf{e}_y + \mathbf{e}_z \right) ,$$
 (3.13)

$$\mathbf{k} = (\bar{1}01) : \mathbf{v} = -\mathbf{e}_y, \quad \mathbf{w} = \frac{1}{\sqrt{1 + (L_x/L_z)^2}} \left((L_x/L_z)\mathbf{e}_x + \mathbf{e}_z \right),$$
 (3.14)

$$\mathbf{k} = (0\bar{1}1)$$
 : $\mathbf{v} = \mathbf{e}_x$, $\mathbf{w} = \frac{1}{\sqrt{1 + (L_x/L_z)^2}} ((L_x/L_z)\mathbf{e}_y + \mathbf{e}_z)$. (3.15)

Finally we need to assign the phase angles $\psi_m(\mathbf{k})$ in a manner that is consistent with the symmetries of the space group $D_4^{10}(I4_122)$. The full list of point symmetries for this space group can be found in the International Tables [86], however, for our purposes we only need to consider the four-fold symmetry about the tetragonal axis and the two-fold symmetry about

the y-axis. The former is a 4₁ screw axis with, in the notation of Eq. (2.44), $\mathbf{t} = (L_z/4)\mathbf{e}_z$ and $\mathbf{y} = (-L_x/4)\mathbf{e}_x + (L_x/4)\mathbf{e}_y$, while the two-fold axis has $\mathbf{t} = \mathbf{0}$ and $\mathbf{y} = (L_x/4)\mathbf{e}_x + (3L_z/8)\mathbf{e}_z$ [86].

Consider first the four-fold axis: under such a transformation $\mathbf{e}_x \to \mathbf{e}_y, \mathbf{e}_y \to -\mathbf{e}_x, \mathbf{e}_z \to \mathbf{e}_z$ and for our choice of gauge $\Theta = 0$. Applying Eq. (2.44) to the (110) and ($\overline{1}10$) wavevectors leads to the relations

$$\psi_m(\bar{1}10) = \psi_m(110) + \pi , \qquad (3.16)$$

$$\psi_m(\bar{1}\bar{1}0) = \psi_m(\bar{1}10) - \pi , \qquad (3.17)$$

from which we immediately deduce that

$$\psi_2(110) = 0, \pi \text{ and } \psi_2(\bar{1}10) = \pi, 0.$$
 (3.18)

Similarly, applying Eq. (2.44) in turn to the $(101), (011), (\bar{1}01)$ and $(0\bar{1}1)$ wavevectors yields the relations

$$\psi_m(011) = \psi_m(101) + \frac{3\pi}{2},$$
(3.19)

$$\psi_m(\bar{1}01) = \psi_m(011) + \frac{\pi}{2} , \qquad (3.20)$$

$$\psi_m(0\bar{1}1) = \psi_m(\bar{1}01) - \frac{\pi}{2} , \qquad (3.21)$$

$$\psi_m(101) = \psi_m(0\bar{1}1) + \frac{\pi}{2} . \tag{3.22}$$

Combining these we may deduce further that $\psi_m(101) = \psi_m(\bar{1}01)$ and $\psi_m(011) = \psi_m(0\bar{1}1)$. Finally, by considering the effect of the two-fold axis on the (101) and (011) wavevectors we obtain the additional relations

$$\psi_m(\bar{1}0\bar{1}) = \psi_m(101) - \frac{\pi}{2},$$
(3.23)

$$\psi_m(01\bar{1}) = \psi_m(011) - \frac{3\pi}{2},$$
(3.24)

from which we conclude that

$$\psi_2(101) = \psi_2(\bar{1}01) = \frac{\pi}{4}, \frac{-3\pi}{4} \text{ and } \psi_2(011) = \psi_2(0\bar{1}1) = \frac{-\pi}{4}, \frac{3\pi}{4}.$$
 (3.25)

It is noteworthy that, as for the space group $O^8(I4_132)$ relevant to blue phase I, there is

more than one choice of the phase angles $\psi_m(\mathbf{k})$ compatible with $D_4^{10}(I4_122)$ symmetry. In this case there are four distinct possibilities since the two choices in Eq. (3.18) can be made independently of the two choices in Eq. (3.25).

With a choice of gauge defined and the Fourier phases determined, a straightforward calculation gives the high chirality form of the \mathbf{Q} -tensor

$$\begin{split} Q_{xx} &= -e^{i\psi_2(110)} \frac{Q_2(110)}{\sqrt{4}} \sin\left(\frac{2\pi}{L_x}(x-x_0)\right) \sin\left(\frac{2\pi}{L_x}(y-y_0)\right) \\ &\quad - \frac{Q_2(101)}{\sqrt{8}} \left[2\cos\left(\frac{2\pi}{L_x}(y-y_0)\right) \sin\left(\frac{2\pi}{L_z}(z-z_0) - \psi_2(101)\right) \right. (3.26) \\ &\quad + \frac{2}{1+(L_x/L_x)^2}\cos\left(\frac{2\pi}{L_x}(x-x_0)\right) \cos\left(\frac{2\pi}{L_z}(z-z_0) - \psi_2(101)\right)\right], \\ Q_{yy} &= -e^{i\psi_2(110)} \frac{Q_2(110)}{\sqrt{4}} \sin\left(\frac{2\pi}{L_x}(x-x_0)\right) \sin\left(\frac{2\pi}{L_x}(y-y_0)\right) \\ &\quad + \frac{Q_2(101)}{\sqrt{8}} \left[2\cos\left(\frac{2\pi}{L_x}(x-x_0)\right) \cos\left(\frac{2\pi}{L_z}(z-z_0) - \psi_2(101)\right) \right], \\ Q_{xy} &= -e^{i\psi_2(110)} \frac{Q_2(110)}{\sqrt{4}} \cos\left(\frac{2\pi}{L_x}(x-x_0)\right) \cos\left(\frac{2\pi}{L_x}(y-y_0)\right) \\ &\quad + \frac{Q_2(101)}{\sqrt{8}} \frac{Q_2(110)}{\sqrt{4}} \cos\left(\frac{2\pi}{L_x}(x-x_0)\right) \cos\left(\frac{2\pi}{L_x}(z-z_0) - \psi_2(101)\right) \\ &\quad - \cos\left(\frac{2\pi}{L_x}(x-x_0)\right) \sin\left(\frac{2\pi}{L_x}(z-z_0) - \psi_2(101)\right) \\ &\quad - \cos\left(\frac{2\pi}{L_x}(x-x_0)\right) \sin\left(\frac{2\pi}{L_x}(x-z_0) - \psi_2(101)\right) \\ &\quad + \frac{Q_2(101)}{\sqrt{8}} \frac{Q_2(110)}{\sqrt{4}} \sqrt{2} \cos\left(\frac{2\pi}{L_x}(x-x_0)\right) \sin\left(\frac{2\pi}{L_x}(y-y_0)\right) \\ &\quad + \frac{Q_2(101)}{\sqrt{8}} \frac{Q_2(110)}{\sqrt{4}} \sqrt{2} \cos\left(\frac{2\pi}{L_x}(x-x_0)\right) \cos\left(\frac{2\pi}{L_x}(z-z_0) - \psi_2(101)\right) \\ &\quad - \frac{1}{\sqrt{1+(L_x/L_x)^2}}} \left[\sin\left(\frac{2\pi}{L_x}(x-x_0)\right) \cos\left(\frac{2\pi}{L_x}(z-z_0) - \psi_2(101)\right)\right], \end{aligned}$$

$$Q_{xz} &= -e^{i\psi_2(110)} \frac{Q_2(110)}{\sqrt{4}} \sqrt{2} \sin\left(\frac{2\pi}{L_x}(x-x_0)\right) \cos\left(\frac{2\pi}{L_x}(z-z_0) - \psi_2(101)\right) \\ &\quad - \frac{1}{\sqrt{1+(L_x/L_x)^2}}} \left[\sin\left(\frac{2\pi}{L_x}(x-x_0)\right) \cos\left(\frac{2\pi}{L_x}(z-z_0) - \psi_2(101)\right)\right], \end{aligned}$$

$$(3.29)$$

$$\left. - \frac{1}{\sqrt{1+(L_x/L_x)^2}} \left[\sin\left(\frac{2\pi}{L_x}(x-x_0)\right) \cos\left(\frac{2\pi}{L_x}(z-z_0) - \psi_2(101)\right)\right], \end{aligned}$$

where (x_0, y_0, z_0) is the location of the origin within the simulation unit cell. The two choices for $\psi_2(110)$ differ only by the overall sign of the $Q_2(110)$ terms. Similarly, the two choices for $\psi_2(101)$ differ only by the overall sign of the $Q_2(101)$ terms. However, the difference between these two choices may equally well be absorbed into a shift of z_0 , so that we do not expect them to lead to distinct blue phase textures. Thus we expect two distinct D_4^{10} textures corresponding to the two choices for $\psi_2(110)$. By analogy with O^8 we denote these as $D_4^{10\pm}$, with the plus sign corresponding to the choice $\psi_2(110) = \pi$.

This is indeed what is found in simulations and the two resulting textures are shown in Fig. 3.5. The D_4^{10+} texture is seen to agree with the disclination network of our candidate blue phase X obtained by switching from blue phase I (Fig. 3.3(II)). Furthermore, in all simulations it was found to have lower free energy than the D_4^{10-} texture under identical conditions. Moreover, the D_4^{10-} texture was found to be less numerically robust than D_4^{10+} . In particular, the form of the disclination network is sensitive to the relative magnitude of the two coefficients $Q_2(110)$ and $Q_2(101)$ appearing in Eq. (3.30). For values of $Q_2(110)$ smaller than $Q_2(101)$ we obtained the disclination network shown in Fig. 3.5, however, for $Q_2(110) > Q_2(101)$ a different, but similar, texture is found with the four-fold symmetry of the tetragonal axis broken to just two-fold (not shown).



Figure 3.5: Textures of the two candidate tetragonal phases; D_4^{10+} (a) and D_4^{10-} (b). The disclinations are shown in blue and the double twist cylinders in grey. The top panel provides a three dimensional view, while the bottom panel shows the projection along the tetragonal axis. 2^3 unit cells have been shown for clarity.

CHAPTER 4

Flexoelectric blue phases

Elastic distortions in the nematic phase of liquid crystals can be categorised into three types: splay, twist and bend. Stable phases with two-dimensional twist distortions, the cholesteric blue phases, have been well characterised and observed experimentally. In this Chapter we describe the occurrence of structures with two-dimensional splay-bend distortions.

When nematics are doped with chiral molecules, the chirality of the dopants imparts a natural twist to the director field. As we have seen in Chapter 2 the texture of these cholesteric phases is generally a *one*-dimensional helix, Fig. 4.1(a). However, phases with two-dimensional helical ordering are also possible, Fig. 4.2(a), and under appropriate conditions some of these textures can be made stable. These are the blue phases of Chapter 2. One might ask whether analogous behaviour could be observed in a liquid crystal which shows splay and bend, rather than twist, distortions.

In 1969 Meyer showed that the one-dimensional splay-bend distortion of the director field, shown in Fig. 4.1(b), could result from flexoelectric coupling to an external electric field [87]. Here we extend these results to show that near the isotropic-nematic transition two-dimensional

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		111/12
	11111	11/1/2
		11/1/
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Figure 4.1: Schematic representation of elastic distortions of nematic liquid crystals, showing the director field projected into the plane of the page. (a) Twist in the cholesteric phase of chiral nematics. (b) Splay and bend in a nematic with flexoelectric interactions.



Figure 4.2: Schematic examples of two-dimensional director field configurations. (a) Structure displaying regions of local double twist, characteristic of cholesteric blue phases. (b) Analogous structure possessing double splay distortions.

splay-bend structures, e.g., Fig. 4.2(b), can be stable. We shall term such phases flexoelectric blue phases.

4.1 Flexoelectricity

The coupling of a liquid crystal to an external field occurs in two principal forms. The most usual is dielectric, where the field couples directly to the orientational order parameter. This coupling is quadratic in the field strength and the molecules tend to align parallel or perpendicular to the field, depending on the sign of the dielectric anisotropy. Flexoelectricity, which we shall deal with here, occurs because of the elastic properties of liquid crystals. If the liquid crystal is composed of pear- or banana-shaped molecules an elastic distortion can lead to a spontaneous polarisation. Conversely, if a polarisation is induced by an external electric field then an elastic distortion results. The polarisation of the liquid crystal reflects the elastic distortion which accompanies it, taking the form $\mathbf{P} = e_{s}\mathbf{n}(\nabla \cdot \mathbf{n}) - e_{b}(\mathbf{n} \cdot \nabla)\mathbf{n}$, and contributing a term $\mathbf{E} \cdot \mathbf{P}$ to the free energy density. This is termed flexoelectric coupling, and it gives a response linear in the field strength [1,87].

Here, we consider a bulk sample of liquid crystal with periodic boundary conditions so that all surface terms may be neglected, further assume that the liquid crystal has zero dielectric anisotropy, so that the effects of the field are purely flexoelectric, and that the electric field inside the liquid crystal is uniform and equal to the applied external field. Flexoelectricity involves the coupling of an electric field to *gradients* in the order parameter, which we briefly review to define our notational conventions. Up to second order in \mathbf{Q} we can construct four invariants of this form:

$$E_{\alpha} \nabla_{\beta} Q_{\alpha\beta} , \qquad (4.1)$$

$$E_{\alpha}Q_{\beta\gamma}\nabla_{\alpha}Q_{\beta\gamma} , \qquad (4.2)$$

$$E_{\alpha}Q_{\alpha\beta}\nabla_{\gamma}Q_{\beta\gamma} , \qquad (4.3)$$

$$E_{\alpha}Q_{\beta\gamma}\nabla_{\beta}Q_{\alpha\gamma}.$$

$$(4.4)$$

The first and second of these are total divergences and hence can be discarded, while any arbitrary linear combination of the last two invariants may be written as

$$A E_{\alpha}(Q_{\alpha\gamma}\nabla_{\gamma}Q_{\beta\gamma} + Q_{\beta\gamma}\nabla_{\beta}Q_{\alpha\gamma}) + B E_{\alpha}(Q_{\alpha\beta}\nabla_{\gamma}Q_{\beta\gamma} - Q_{\beta\gamma}\nabla_{\beta}Q_{\alpha\gamma}).$$
(4.5)

The first of these terms is a total divergence, $E_{\alpha} \nabla_{\gamma} (Q_{\alpha\beta} Q_{\beta\gamma})$, and therefore represents only a surface electric field coupling. Hence we find that bulk flexoelectricity is given by the unique coupling

$$F_{\text{flexo}} = \frac{1}{V} \int_{\Omega} \mathrm{d}^3 r \; Q_{\alpha\beta} \big[\varepsilon_{\text{f}} (E_{\alpha} \nabla_{\gamma} - E_{\gamma} \nabla_{\alpha}) \delta_{\beta\delta} \big] Q_{\gamma\delta} \;, \tag{4.6}$$

where V is the volume of the domain, Ω . Analogous arguments have been made some time ago for flexoelectric coupling expressed in terms of the director field [88]. This is not to say that the surface terms neglected above are always unimportant. Indeed quite the converse is true in many cases, particularly when the surface displays topological patterning [89–93]. Nonetheless, here we confine our attention to purely bulk effects and henceforth neglect all such surface terms.

4.2 Landau-de Gennes theory

The Landau-de Gennes free energy in the presence of both chiral and flexoelectric couplings is

$$f = \frac{1}{V} \int_{\Omega} d^3 r \left\{ \frac{\tau}{4} \operatorname{tr} \left(\mathbf{Q}^2 \right) - \sqrt{6} \operatorname{tr} \left(\mathbf{Q}^3 \right) + \left(\operatorname{tr} \left(\mathbf{Q}^2 \right) \right)^2 + \frac{27L_1}{4A_0 \gamma} \left[\left(\nabla \mathbf{Q} \right)^2 + 4q_0 \mathbf{Q} \cdot \nabla \times \mathbf{Q} + \frac{2\varepsilon_{\mathrm{f}}}{L_1} Q_{\alpha\beta} \left(E_\alpha \nabla_\gamma - E_\gamma \nabla_\alpha \right) Q_{\beta\gamma} \right] \right\},$$

$$(4.7)$$

where we have adopted a dimensionless form for the free energy in a similar manner to that for cholesteric liquid crystals in Chapter 2. For simplicity, we adopt the one elastic constant approximation, where the magnitude of twist, splay and bend are taken to be equal. If we take the view that flexoelectricity acts to induce structure in the liquid crystal in an analogous way to chirality, then it is natural to again seek stable states by introducing a Fourier decompositon for \mathbf{Q} , Eq. (2.41). When this is done the quadratic part of the free energy takes the form

$$f^{(2)} = \sum_{\mathbf{k}} N_k^{-1} \sum_{m_1, m_2} Q_{m_1}(k) Q_{m_2}(k) e^{i(\psi_{m_2}(\mathbf{k}) - \psi_{m_1}(\mathbf{k}))} M_{m_1}^{\dagger}(\hat{\mathbf{k}})_{\alpha\beta} \left\{ \frac{\tau}{4} \delta_{\alpha\gamma} + \frac{27L_1}{4A_0\gamma} \left[k^2 \delta_{\alpha\gamma} - i \, 4q_0 \, k \, \epsilon_{\alpha\delta\gamma} \hat{k}_{\delta} - i \, \frac{2\varepsilon_{\mathrm{f}} E}{L_1} \, k \left(\hat{E}_{\alpha} \hat{k}_{\gamma} - \hat{k}_{\alpha} \hat{E}_{\gamma} \right) \right] \right\} M_{m_2}(\hat{\mathbf{k}})_{\beta\gamma} \,.$$

$$(4.8)$$

The basis tensors $\mathbf{M}_m(\hat{\mathbf{k}})$ should be chosen so as to diagonalise $f^{(2)}$ as was done for purely chiral coupling [60]. It is convenient to introduce the quantity

$$\Delta_{\mathbf{k}} \coloneqq \left[1 - \left(\hat{\mathbf{E}}.\hat{\mathbf{k}}\right)^2\right]^{1/2},\tag{4.9}$$

and use the direction of the electric field and of the wavevector for a given Fourier mode to define a local, right-handed, orthonormal frame field $\{\hat{\mathbf{k}}, \mathbf{v}, \mathbf{w}\}$ such that $\hat{\mathbf{E}} = (\hat{\mathbf{E}}.\hat{\mathbf{k}})\hat{\mathbf{k}} + \Delta_{\mathbf{k}}\mathbf{w}$. In terms of this local frame the flexoelectric coupling then takes the form $+2ik(\varepsilon_{\mathrm{f}}E\Delta_{\mathbf{k}}/L_{1})\epsilon_{\alpha\delta\gamma}v_{\delta}$ revealing a formal mathematical similarity between flexoelectric and chiral couplings. Combining the two terms motivates a transformation to a new set of basis vectors, obtained by means of a rotation about \mathbf{w} :

$$\mathbf{a} := \frac{1}{\mu_{\mathbf{k}}} \left(2q_0 \hat{\mathbf{k}} - \frac{\varepsilon_{\mathrm{f}} E \Delta_{\mathbf{k}}}{L_1} \mathbf{v} \right) ,$$

$$\mathbf{b} := \frac{1}{\mu_{\mathbf{k}}} \left(\frac{\varepsilon_{\mathrm{f}} E \Delta_{\mathbf{k}}}{L_1} \hat{\mathbf{k}} + 2q_0 \mathbf{v} \right) ,$$

$$\mathbf{c} := \mathbf{w} ,$$
(4.10)

where

$$\mu_{\mathbf{k}} = \left[\left(2q_0 \right)^2 + \left(\frac{\varepsilon_{\mathbf{f}} E \Delta_{\mathbf{k}}}{L_1} \right)^2 \right]^{1/2}, \qquad (4.11)$$

is a normalisation factor that also serves to define a natural length scale, generalising $2q_0$ in the purely chiral case. In this rotated local frame, the choice of orthonormal basis tensors

$$\mathbf{M}_{\pm 2} = \frac{1}{2} (\mathbf{b} \pm i\mathbf{c}) \otimes (\mathbf{b} \pm i\mathbf{c}) ,$$

$$\mathbf{M}_{\pm 1} = \frac{1}{2} \Big[\mathbf{a} \otimes (\mathbf{b} \pm i\mathbf{c}) + (\mathbf{b} \pm i\mathbf{c}) \otimes \mathbf{a} \Big] ,$$

$$\mathbf{M}_{0} = \frac{1}{\sqrt{6}} (3\mathbf{a} \otimes \mathbf{a} - \mathbf{I}) ,$$

(4.12)

results in the desired diagonalisation of $f^{(2)}$,

$$f^{(2)} = \sum_{\mathbf{k}} N_k^{-1} \sum_{m=-2}^2 Q_m^2(k) \left\{ \frac{\tau}{4} + \frac{27L_1}{4A_0\gamma} \left[k^2 - m\mu_{\mathbf{k}} k \right] \right\}.$$
 (4.13)

We see from Eqs. (4.10) that there is strong sense in which the transition from purely chiral to purely flexoelectric coupling may be viewed geometrically as a $\pi/2$ rotation. Indeed, this is already evident in the one-dimensional cholesteric and splay-bend states of Fig. 4.1, which are related by a $\pi/2$ rotation about the vertical direction. This rotation is none other than the well known flexoelectro-optic effect [94].

4.3 Flexoelectric blue phases

Given this formal similarity it is natural to ask whether a precise correspondence can be made between the stable phases, and their properties, in the purely chiral and purely flexoelectric limits. To this end we now focus our attention on the purely flexoelectric sector and analyse a number of structures motivated by the analogy with cholesterics.

The simplest flexoelectric structure is a one-dimensional phase that was described in Meyer's original Letter [87], which we shall call splay–bend; see Fig. 4.1(b). The **Q**-tensor appropriate to splay–bend is the analogue of that for the cholesteric helix in chiral liquid crystals:

$$\mathbf{Q} = \frac{Q_2}{\sqrt{2}} \begin{pmatrix} \cos(kx) & 0 & \sin(kx) \\ 0 & 0 & 0 \\ \sin(kx) & 0 & -\cos(kx) \end{pmatrix} - \frac{Q_h}{\sqrt{6}} \begin{pmatrix} -1 & 0 & 0 \\ 0 & 2 & 0 \\ 0 & 0 & -1 \end{pmatrix} , \qquad (4.14)$$

where we have used the electric field to define the z-axis of a Cartesian coordinate system and taken the wavevector to be orthogonal to the field so as to maximise $\Delta_{\mathbf{k}}$ and hence minimise the free energy. The free energy is readily shown to be

$$f = \frac{\tau}{4} \left(Q_2^2 + Q_h^2 \right) - 3Q_2^2 Q_h + Q_h^3 + \left(Q_2^2 + Q_h^2 \right)^2 + \frac{27L_1}{4A_0\gamma} Q_2^2 \left(k^2 - 2\frac{\varepsilon_{\rm f}E}{L_1} k \right) \,. \tag{4.15}$$

It is formally identical to that of the cholesteric phase with $\varepsilon_{\rm f} E/L_1$ playing the role of the chiral parameter, $2q_0$. Therefore the solutions for Q_2 and Q_h as a function of the reduced temperature, τ , and field strength, E, obtained by minimising Eq. (4.15), are the same as those obtained for the cholesteric phase [42,60,82] (Eqs. (2.32) and (2.33)), provided one substitutes $2q_0$ with $\varepsilon_{\rm f} E/L_1$.

Extending the analogy with cholesterics, we now consider flexoelectric phases with a higher dimensional structure; flexoelectric analogues of the cholesteric blue phases. Based on the heuristic observations that the field picks out a preferred direction in space, and that the flexoelectric coupling vanishes for wavevectors parallel to the field, it seems likely that the free energy will be minimised by two-dimensional structures, containing only wavevectors orthogonal to the field. Therefore we consider first a two-dimensional phase with hexagonal symmetry, obtained by choosing the fundamental set of Fourier modes to be along the directions $\{\pm \mathbf{e}_x, \pm (\mathbf{e}_x + \sqrt{3}\mathbf{e}_y)/2, \pm (\mathbf{e}_x - \sqrt{3}\mathbf{e}_y)/2\}$. An approximate **Q**-tensor, comprising only this fundamental set, all with m = 2, and a homogeneous component, is given by

$$\mathbf{Q} = \frac{Q_2}{\sqrt{6}} \left\{ \frac{1}{2} \left(\mathbf{e}_x + i\mathbf{e}_z \right) \otimes^S e^{-i(kx - \psi_1)} + \frac{1}{2} \left(\frac{-1}{2} \mathbf{e}_x + \frac{\sqrt{3}}{2} \mathbf{e}_y + i\mathbf{e}_z \right) \otimes^S e^{-i(k(-x + \sqrt{3}y)/2 - \psi_2)} \right. \\ \left. + \frac{1}{2} \left(\frac{-1}{2} \mathbf{e}_x - \frac{\sqrt{3}}{2} \mathbf{e}_y + i\mathbf{e}_z \right) \otimes^S e^{-i(k(-x - \sqrt{3}y)/2 - \psi_3)} + \text{ H.c.} \right\} + \frac{Q_h e^{i\delta}}{\sqrt{6}} \left(3\mathbf{e}_z \otimes \mathbf{e}_z - \mathbf{I} \right) ,$$

$$(4.16)$$

where H.c. stands for Hermitian conjugate, $\psi_1, \psi_2, \psi_3 \in [0, 2\pi)$, $\delta \in \{0, \pi\}$ and the short-hand notation \otimes^S denotes a symmetrised tensor product, i.e., $\mathbf{u} \otimes^S := \mathbf{u} \otimes \mathbf{u}$. The director field of this hexagonal flexoelectric blue phase is shown in Fig. 4.3. The structure consists of an hexagonal lattice of strength -1/2 disclinations separating regions in which the distortion is one of pure splay along any straight line passing through the centre of the hexagon. Indeed, expanding in cylindrical polars (ρ, ϕ, z), the director field in a local neighbourhood of the centre of each hexagon is found to be

$$\mathbf{n} = \cos\left(\frac{Q_2 k\rho}{3Q_2 + 2Q_h}\right) \mathbf{e}_z - \sin\left(\frac{Q_2 k\rho}{3Q_2 + 2Q_h}\right) \mathbf{e}_\rho , \qquad (4.17)$$



Figure 4.3: Director field of the hexagonal flexoelectric blue phase described in the main text obtained from a numerical minimisation at paramter values $\kappa_{\rm E} = 1, \tau - \kappa_{\rm E}^2 = 0$ where the phase is stable, see Fig. 4.4. The structure is periodic in both directions.

and therefore the structure may be apply referred to as a *double splay cylinder* since it is the clear analogue of double twist cylinders in the cholesteric blue phases.

The free energy of this hexagonal phase

$$f = \frac{\tau}{4} \left(Q_2^2 + Q_h^2 \right) - \frac{\kappa_{\rm E}^2}{4} Q_2^2 - \frac{3e^{i\delta}}{2} Q_2^2 Q_h - e^{i\delta} Q_h^3 + \frac{27}{32} \cos(\psi_1 + \psi_2 + \psi_3) Q_2^3 + \frac{233}{192} Q_2^4 + \frac{7}{2} Q_2^2 Q_h^2 + Q_h^4 - \frac{9}{8} \cos(\psi_1 + \psi_2 + \psi_3 + \delta) Q_2^3 Q_h ,$$

$$(4.18)$$

where we have defined a flexoelectric analogue of the chirality

$$\kappa_{\rm E}^2 \coloneqq \frac{27\varepsilon_{\rm f}^2 E^2}{A_0 \gamma L_1} , \qquad (4.19)$$

is minimised by choosing the phases to satisfy $\psi_1 + \psi_2 + \psi_3 = \pi$ and $\delta = 0$. As for the splay-bend phase, this free energy of the flexoelectric phase is identical to the free energy of its cholesteric analogue, in this case the planar hexagonal blue phase [42, 60]. Consequently, we may draw on this previous work for cholesterics to conclude that the hexagonal flexoelectric phase has lower free energy than the splay-bend phase at sufficiently large field strength. However, in the chiral case the hexagonal blue phase is not stable since one of the cubic blue phases always has lower free energy. It is therefore necessary to see whether a similar result also holds in the flexoelectric case.

Although one may expect that the structure of the cubic blue phases provides a good

starting point for a consideration of flexoelectric blue phases, the latter differ in a number of respects. In particular, since the flexoelectric coupling is vectorial, it defines a preferred direction which will act to lower the symmetry from cubic to tetragonal. We therefore consider the set of possible structures obtained by selecting the Fourier modes of \mathbf{Q} to be

$$\left\{ \mathbf{0}, \pm \frac{2\pi}{L_x} \mathbf{e}_x, \pm \frac{2\pi}{L_x} \mathbf{e}_y, \pm \frac{2\pi}{L_x} \left(\mathbf{e}_x + \mathbf{e}_y \right), \pm \frac{2\pi}{L_x} \left(-\mathbf{e}_x + \mathbf{e}_y \right), \pm \frac{2\pi}{L_z} \mathbf{e}_z, \\ \pm \frac{2\pi}{L_x} \left(\mathbf{e}_x + \frac{L_x}{L_z} \mathbf{e}_z \right), \pm \frac{2\pi}{L_x} \left(\mathbf{e}_y + \frac{L_x}{L_z} \mathbf{e}_z \right), \pm \frac{2\pi}{L_x} \left(-\mathbf{e}_x + \frac{L_x}{L_z} \mathbf{e}_z \right), \pm \frac{2\pi}{L_x} \left(-\mathbf{e}_y + \frac{L_x}{L_z} \mathbf{e}_z \right) \right\},$$
(4.20)

where L_x, L_z are the lattice parameters along the x- and z-axes of the conventional unit cell respectively. This set of structures allows for considerable scope, containing as special cases **Q**-tensors that are exact analogues of those representing the cholesteric cubic blue phases as well as that of the two-dimensional square structure shown in Fig. 4.2(b). We have investigated the free energy minima of these structures both using approximate analytic calculations and with an exact numerical minimisation for a range of initial conditions, varying the ratio of the lattice parameters L_x/L_z and the relative amplitudes and phases of the Fourier components. The numerical minimisation was performed using a lattice Boltzmann algorithm [32,34] with an identical technique to that described in Chapter 2. In all instances a single minimum was obtained that corresponded to the two-dimensional square flexoelectric blue phase of Fig. 4.2(b), a result that further supports our heuristic remarks that the suppression of wavevectors parallel to the field might favour two-dimensional textures.

These results were combined with the analytic expressions for the free energy of the splaybend and hexagonal structures, Eqs. (4.15) and (4.18), to construct an approximate phase diagram. We find that although the two-dimensional square phase is a local minimum of the free energy within the set of structures defined by Eq. (4.20), it never has lower free energy than the hexagonal phase. The same result is obtained numerically, leading to the phase diagram shown in Fig. 4.4, which has been plotted in the $\kappa_{\rm E}$, $(\tau - \kappa_{\rm E}^2)$ plane, in accordance with the conventions adopted in cholesterics [82], see Chapter 2. As the electric field strength is increased, the hexagonal flexoelectric blue phase becomes stabilised over an increasing temperature interval between the isotropic and splay-bend phases. The discrepancy between the numerical and analytic phase boundaries is similar to that found in cholesterics and arises from neglecting higher order wavevector harmonics in analytic calculations. In the cholesteric blue phases the lattice periodicity is not identical to the cholesteric pitch, but is generally somewhat larger.



Figure 4.4: Phase diagram of the flexoelectric blue phases. The circles and solid lines show the numerical phase boundaries, while the analytic results are given by the dashed lines.

From the numerical minimisation we find the same feature in the hexagonal flexoelectric blue phase, with the lattice parameter being approximately 15% larger than the 'pitch' $(\pi L_1/\varepsilon_f E)$ of the splay-bend phase at the triple point.

The phase diagram bears a qualitative resemblance to that of cholesterics (Chapter 2), but the details reveal differences between chiral and flexoelectric couplings. In particular, it seems that the suppression of wavevectors parallel to the field introduced by the quantity $\Delta_{\mathbf{k}}$ is sufficient to convert the fully three-dimensional states which are stable at large chiral coupling into two-dimensional states under flexoelectric coupling.

We comment that the local director field in the double splay regions of the flexoelectric blue phases is the same as that in the escape configuration lattice phase recently suggested by Chakrabarti *et al.* [8]. However, the mechanism by which the structure occurs, and is stabilised, is different. Their proposed structure arises without the application of an electric field, being instead stabilised by the saddle splay elastic constant and including weak anchoring of the director field and surface tension at the interface between the nematic and isotropic regions of the fluid. Note also that the hexagonal symmetry of the phase we obtain is different to the square symmetry predicted in that work.

4.3.1 Experimental estimates

It is important for potential experimental work to estimate the field strength required to stabilise the two-dimensional hexagonal flexoelectric blue phase. A good estimate is that the strength of the flexoelectric coupling, $\varepsilon_{\rm f}E$, should be as large as the chiral coupling, $2q_0L_1$, in a compound which possesses cholesteric blue phases. This gives $E \approx 2q_0L_1/\varepsilon_{\rm f} \approx \pi K/ep$, where we have replaced the Landau–de Gennes parameters with their director field equivalents; an average Frank elastic constant, K, and the average of the flexoelectric coupling constants, $e := (e_{\rm s} + e_{\rm b})/2$. p is the pitch in the cholesteric phase of a compound which also displays blue phases, typically $\sim 0.3\mu m$. Recently materials have been developed for flexoelectric purposes and found to have a large flexoelastic ratio, $e/K \sim 1V^{-1}$ [95]. Thus in these materials the required field strength is expected to be $E \sim 10V\mu m^{-1}$. It will also be helpful to use a material with close to zero dielectric anisotropy and to take care to ensure that surface effects are negligible compared to the bulk, as we have assumed here. The analogy with cholesterics further suggests that flexoelectric blue phases are only likely to be found in a narrow temperature range (~ 1 K) just below the isotropic-nematic transition temperature, although this is predicted to expand with increasing field strength.

4.4 Flexoelectric effects in the cholesteric blue phases

The Landau-de Gennes theory we developed in § 4.2 showed that chiral and flexoelectric couplings could be described within a common framework, the transition from one limiting case to the other occuring via a $\pi/2$ rotation. For one-dimensional textures the cross-over from the cholesteric helix to the flexoelectric splay-bend is smooth, without any kind of sharp transition. However, this smooth cross-over does not extend to the blue phases, for on the chiral side the stable textures are three-dimensional, while on the flexoelectric side they are two-dimensional. Thus at some intermediate stage a transition must occur. In this final section we illustrate this transition for two specific cases: first we will look at blue phase I with a field applied parallel to the [011] direction, and then we will consider blue phase II with a field applied parallel to [001], thereby mirroring our analysis of dielectric switching in Chapter 3. We begin with a brief review of the cross-over between one-dimensional textures.

4.4.1 Flexoelectro-optic effect in cholesterics

If a cholesteric liquid crystal is prepared with its helical axis parallel to the cell plates (uniform lying helix) the cell appears uniformly dark when viewed between crossed polarisers. The cell can be made to transmit light by applying a small electric field perpendicular to the helical axis, indicating a rotation of the optical axis, and extinction subsequently regained by a small rotation of the cell. The rotation angle needed to regain extinction is found to be linear in the applied field strength, for small fields, and to change sign when the direction of the field is reversed. This effect, known as the flexoelectro-optic effect, was first described by Patel and Meyer [94] and has since been developed for potential display technologies [95–97].

If we assume a minimal modification for the **Q**-tensor, then the cholesteric under an applied field will be described by

$$\mathbf{Q} = \frac{Q_2}{\sqrt{2}} \Big[\left(\mathbf{b} \otimes \mathbf{b} - \mathbf{e}_z \otimes \mathbf{e}_z \right) \cos(kx) + \left(\mathbf{b} \otimes \mathbf{e}_z + \mathbf{e}_z \otimes \mathbf{b} \right) \sin(kx) \Big] - \frac{Q_h}{\sqrt{6}} \Big[3\mathbf{a} \otimes \mathbf{a} - \mathbf{I} \Big] , \quad (4.21)$$

where we have taken the helical axis to define the x-axis and then the direction of the electric field to define the xz-plane. This **Q**-tensor provides a homotopy between the zero field, cholesteric (Eq. (2.25)), and the zero chirality, splay-bend (Eq. (4.14)), textures. As such, the effect of flexoelectric coupling on the cholesteric phase is to rotate an initially chiral texture into an achiral one. The angle of rotation, as a function of field strength, is given by

$$\tan(\phi) = \frac{\varepsilon_{\rm f} E \Delta_{\rm k}}{2q_0 L_1} , \qquad (4.22)$$

in agreement with the result first obtained in Reference [94]. This rotation angle provides both a simple, geometrical view of the distortion brought about by the field and also a natural, and convenient, dimensionless measure of the relative strength of the chiral and flexoelectric couplings. Recently rotation angles of as much as 87° have been reported [95].

4.4.2 Blue phase I

When an electric field is applied to a cholesteric blue phase it leads to both a distortion of the director field of the texture and also to a change in the size and shape of the unit cell. At large field strengths the distortion and change in shape can be appreciable, ultimately leading to field-induced textural transitions. For the same reasons as discussed in Chapter 3, the change in size and shape is quadratic in the field strength at low fields and may be described by a change in the lattice vectors of the unit cell through a fourth rank electrostriction tensor. However, unlike the analogous case of dielectric coupling, nothing is currently known about the electrostriction tensor arising from flexoelectric effects: neither the typical magnitude, nor signs of R_1 , R_2 and



Figure 4.5: Flexoelectric switching of blue phase I for fields parallel to [011]. At small field strengths the unit cell distorts to become tetragonal, resulting in a textural transition to a centred tetragonal phase with the same space group as blue phase X (see § 3.3). At larger field strengths a second transition is found to a simple tetragonal phase with space group $P4_222$. At still larger fields there is a final transition to the two-dimensional hexagonal flexoelectric blue phase. The disclination lines are shown in blue and 2^3 unit cells have been shown for clarity.

 R_3 . Nonetheless, the eventual transition to the two-dimensional hexagonal flexoelectric blue phase and suppression of wavevectors parallel to the field brought about by the quantity $\Delta_{\mathbf{k}}$ suggest that the texture should undergo an expansion parallel to the field direction, in which case both R_1 and R_3 would be positive. Preliminary numerical simulations indicate that this is the case and that when the field is applied parallel to the crystallographic [011] direction, it promotes a distortion towards a tetragonal geometry, as is also found under dielectric coupling. Therefore, this is the geometry we employ to look at the chiral-to-flexoelectric cross-over in blue phase I.

Blue phase I is initialised in a tetragonal unit cell of size $32 \times 32 \times 40$ with its [011] direction parallel to the z-axis of the simulation unit cell. The texture is allowed to relax for a short period before an electric field is applied parallel to the z-axis. As was found for dielectric effects in Chapter 3, the electric field induces a twist in the disclination lines allowing them to transiently merge, thus fascilitating textural transitions. These are illustrated in Fig. 4.5, with the field strength at which the transitions occur quoted in terms of $tan(\phi)$, the dimensionless measure of the relative strength of chiral and flexoelectric couplings. For the largest applied field strengths a transition is indeed observed to the two-dimensional hexagonal flexoelectric



Figure 4.6: Flexoelectric switching of blue phase II for fields parallel to [001]. The disclination lines are shown in blue and the double twist cylinders in grey. In all cases 2^3 unit cells have been shown for clarity. For the hexagonal flexoelectric blue phase we show the director field configuration viewed along the field direction and show 2^2 simulation unit cells. The hexagonal symmetry of the preferred texture is evident despite the imposed square symmetry of the simulation box.

blue phase, as expected. However, at intermediate values, two further transitions precede it, each yielding distinct textures that are stable over a small range of field strengths. The first it to a centred tetragonal texture with space group $I4_122$, which we have called blue phase F_I and that we recognise as having the same disclination network as the blue phase X structure found under dielectric switching and described in § 3.3.1. At slightly higher field strengths there is a second transition to a distinct tetragonal texture that we have called blue phase F_{II} , this time with space group $P4_222$. In this texture, the disclinations are all parallel to the field, occuring in pairs that wrap around each other to form a double helix, and with the axes of the double helices themselves then arranged on a square lattice. Finally, the two members of a given double helix transiently merge and re-separate, allowing them to both straighten out and to adopt an hexagonal configuration in the plane perpendicular to the field.

4.4.3 Blue phase II

We now follow a similar analysis, but for blue phase II and with the field applied parallel to the crystallographic [001] direction. The sequence of transitions is shown in Fig. 4.6. At relatively small field strengths there is an unbinding of the bicontinuous network of disclinations, in a
similar manner to that found under dielectric coupling in § 3.2, but with the unbinding occuring in a different direction. This is accompanied by a breaking of the double twist cylinders which lie initially perpendicular to the field. After this unbinding the disclinations form pairs of double helices arranged on a square lattice, which we identify with the texture blue phase F_{II} also found in the switching of blue phase I. Finally, at field strengths of $tan(\phi) \sim 1.4$ there is a further transition to a two-dimensional texture corresponding to the hexagonal flexoelectric blue phase.

Double twist cylinders

We conclude this Chapter with a brief analysis of the small field distortion of blue phase II, focusing on the changes induced in the double twist cylinders. We have seen in § 2.5 that an approximate \mathbf{Q} -tensor containing only the fundamental set of wavevectors is sufficient to correctly describe the blue phase texture. For small field strengths we can assume that the principal effects of the electric field are captured by the same approximate \mathbf{Q} -tensor, but with the basis tensors of Eq. (4.12), which are appropriate to flexoelectric coupling,

$$\mathbf{Q} = \frac{Q_{2}(k_{\perp})}{\sqrt{4}} \begin{pmatrix} \left(\frac{\varepsilon_{f}E}{\mu L_{1}}\right)^{2} \cos(k_{\perp}x) & \frac{2q_{0}\varepsilon_{f}E}{\mu^{2}L_{1}} \cos(k_{\perp}x) & \frac{\varepsilon_{f}E}{\mu L_{1}} \sin(k_{\perp}x) \\ \frac{2q_{0}\varepsilon_{f}E}{\mu^{2}L_{1}} \cos(k_{\perp}x) & \left(\frac{2q_{0}}{\mu}\right)^{2} \cos(k_{\perp}x) & \frac{2q_{0}}{\mu} \sin(k_{\perp}x) \\ \frac{\varepsilon_{f}E}{\mu L_{1}} \sin(k_{\perp}x) & \frac{2q_{0}}{\mu} \sin(k_{\perp}x) & -\cos(k_{\perp}x) \end{pmatrix} \\ - \frac{Q_{2}(k_{\perp})}{\sqrt{4}} \begin{pmatrix} \left(\frac{2q_{0}}{\mu}\right)^{2} \cos(k_{\perp}y) & \frac{-2q_{0}\varepsilon_{f}E}{\mu^{2}L_{1}} \cos(k_{\perp}y) & \frac{-2q_{0}}{\mu} \sin(k_{\perp}y) \\ \frac{-2q_{0}\varepsilon_{f}E}{\mu^{2}L_{1}} \cos(k_{\perp}y) & \left(\frac{\varepsilon_{f}E}{\mu L_{1}}\right)^{2} \cos(k_{\perp}y) & \frac{\varepsilon_{f}E}{\mu L_{1}} \sin(k_{\perp}y) \\ \frac{-2q_{0}}{\mu} \sin(k_{\perp}y) & \frac{\varepsilon_{f}E}{\mu L_{1}} \sin(k_{\perp}y) & -\cos(k_{\perp}y) \end{pmatrix} \\ + \frac{Q_{2}(k_{\parallel})}{\sqrt{2}} \begin{pmatrix} \cos(k_{\parallel}z) & \sin(k_{\parallel}z) & 0 \\ \sin(k_{\parallel}z) & -\cos(k_{\parallel}z) & 0 \\ 0 & 0 & 0 \end{pmatrix} . \end{cases}$$

$$(4.23)$$

Here, we have also allowed for the electric field to lower the symmetry from cubic to tetragonal by assigning different wavevector magnitudes and Fourier amplitudes to the modes parallel and perpendicular to the field. The limit $E \to 0$ is recovered by requiring $k_{\perp} \to k_{\parallel}$ and $Q_2(k_{\perp}) \to \sqrt{2}Q_2(k_{\parallel})$.

In the cholesteric texture, the effect of the electric field could be simply interpreted as a rotation of the director about the z-axis. We shall show that the distortion of the double twist

cylinders in blue phase II may be similarly described in terms of a rotation of the director. Two distinct cases need to be considered: cylinders whose axes are initially parallel to the field, and cylinders whose axes are initially perpendicular to the field. Starting with the former case, we can expand the **Q**-tensor about the line (1/2, 0, z) to obtain

$$\mathbf{Q} = \frac{Q_2(k_{\perp})}{\sqrt{4}} \begin{pmatrix} -1 & 0 & -\frac{\varepsilon_{\mathrm{f}} E k_{\perp} \delta x}{\mu L_1} + \frac{2q_0 k_{\perp} \delta y}{\mu} \\ 0 & -1 & -\frac{\varepsilon_{\mathrm{f}} E k_{\perp} \delta y}{\mu L_1} - \frac{2q_0 k_{\perp} \delta x}{\mu L_1} \\ -\frac{\varepsilon_{\mathrm{f}} E k_{\perp} \delta x}{\mu L_1} + \frac{2q_0 k_{\perp} \delta y}{\mu} & -\frac{\varepsilon_{\mathrm{f}} E k_{\perp} \delta y}{\mu L_1} - \frac{2q_0 k_{\perp} \delta x}{\mu} & 2 \end{pmatrix}$$

$$+ \frac{Q_2(k_{\parallel})}{\sqrt{2}} \begin{pmatrix} \cos(k_{\parallel} z) & \sin(k_{\parallel} z) & 0 \\ \sin(k_{\parallel} z) & -\cos(k_{\parallel} z) & 0 \\ 0 & 0 & 0 \end{pmatrix}.$$

$$(4.24)$$

If we similarly expand the director in a neighbourhood of the cylinder axis as $\mathbf{n} = (n_x, n_y, 1)$ then a short calculation reveals that

$$\begin{pmatrix} n_x \\ n_y \end{pmatrix} = \frac{Q_2(k_\perp)}{9Q_2^2(k_\perp) - 2Q_2^2(k_\parallel)} \begin{pmatrix} -\sqrt{2}Q_2(k_\parallel)\sin(k_\parallel z) & 3Q_2(k_\perp) + \sqrt{2}Q_2(k_\parallel)\cos(k_\parallel z) \\ -3Q_2(k_\perp) + \sqrt{2}Q_2(k_\parallel)\cos(k_\parallel z) & \sqrt{2}Q_2(k_\parallel)\sin(k_\parallel z) \end{pmatrix} \times \begin{pmatrix} \frac{2q_0}{\mu} & \frac{\varepsilon_f E}{\mu L_1} \\ \frac{-\varepsilon_f E}{\mu L_1} & \frac{2q_0}{\mu} \end{pmatrix} \begin{pmatrix} k_\perp \delta x \\ k_\perp \delta y \end{pmatrix} .$$

$$(4.25)$$

Thus the distortion of the cylinder can indeed be viewed as a rotation about the z-axis, again through an angle $\phi = \arctan(\varepsilon_{\rm f} E/2q_0 L_1)$, as in the cholesteric flexoelectro-optic effect. However, whereas for the cholesteric the distorted director is obtained by rotating the local director without displacing it from its initial position, we see here that the distorted double twist cylinder at a point $\delta \mathbf{r} := (\delta x, \delta y)$ is given by the undistorted configuration at the rotated point $\mathbf{R}\delta \mathbf{r}$, with \mathbf{R} a rotation matrix through angle ϕ . Thus the effect of the field is to induce a bulk rotation of the entire double twist cylinder.

The situation turns out to be quite different for cylinders whose axes are initially perpendicular to the field. One such axis, described in § 2.5, is the line (x, 1/2, 0), defined by the locus of points for which the director is $\mathbf{n} = (1, 0, 0)$. In non-zero field one may readily verify that the line (x, 1/2, 0) no longer describes the cylinder axis, which instead is given by solutions of the simultaneous equations

$$\frac{Q_2(k_{\perp})}{2} \left(\frac{\varepsilon_{\rm f} E}{\mu L_1}\right)^2 \cos(k_{\perp} x) - \frac{Q_2(k_{\perp})}{2} \left(\frac{2q_0}{\mu}\right)^2 \cos(k_{\perp} y) + \frac{Q_2(k_{\parallel})}{\sqrt{2}} \cos(k_{\parallel} z) = S , \qquad (4.26)$$

$$\frac{Q_2(k_{\perp})}{2} \frac{2q_0 \varepsilon_{\rm f} E}{\mu^2 L_1} \left(\cos(k_{\perp} x) + \cos(k_{\perp} y) \right) + \frac{Q_2(k_{\parallel})}{\sqrt{2}} \sin(k_{\parallel} z) = 0 , \qquad (4.27)$$

$$\frac{\varepsilon_{\rm f}E}{\mu L_1}\sin(k_{\perp}x) + \frac{2q_0}{\mu}\sin(k_{\perp}y) = 0 , \qquad (4.28)$$

where S is the maximal eigenvalue of \mathbf{Q} . The last of these equations may be simply rearranged to give

$$\sin(k_{\perp}y) = -\tan(\phi)\sin(k_{\perp}x) . \tag{4.29}$$

This is an important result, which shows that the axis only remains continuous as one moves along the x-direction if $tan(\phi) < 1$, i.e., so long as the chiral coupling remains larger than the flexoelectric coupling. For field strengths above this value the cylinders perpendicular to the field must necessarily break. Of course, this estimate provides only an upper bound and it is perfectly plausible that the cylinders will break at much lower field strengths.

For small field strengths, $\tan(\phi) \ll 1$, we can expand about the zero field solution, letting the cylinder axis be given by $(x, 1/2 + \delta y, \delta z)$, whereupon we find

$$\delta y = \frac{1}{2\pi} \tan(\phi) \sin(k_\perp x) , \qquad (4.30)$$

$$\delta z = \frac{1}{\pi} \tan(\phi) \sin^2(k_{\perp} x/2) .$$
(4.31)

Note that while the displacement perpendicular to the field, δy , averages to zero over the unit cell, the displacement parallel to the field, δz , does not. We might say that the mean position of the cylinder has been raised in the direction parallel to the field by an amount linearly proportional to the field strength, at least for small fields. This mean position is given by the line $(x, 1/2, (1/2\pi) \tan(\phi))$, which the cylinder axis wraps around in a helical fashion.

The distortion and breaking of double twist cylinders that we have just described is in good qualitative agreement with the results of a full numerical simulation shown in Fig. 4.6.

CHAPTER 5

Locomotion: low Reynolds number swimmers

The ability to move is a ubiquitous trait. From birds in the sky to wildebeast on the plains of the Serengeti and cyclists on the streets of Oxford, living things are able to generate their own motion. Understanding this motion has long been a topic of interest to biologists, mechanical engineers and applied mathematicians, but recently it has also attracted growing interest from condensed matter physicists, primarily because of the remarkable properties of collective locomotion in flocks and herds [98].

Systems as seemingly unrelated as flocks of birds, swarms of bacteria and layers of vibrated granular rods all display features of long, or quasi-long, range order and large scale hydrodynamic swirls and jets, leading to their continuum description within the common theoretical framework of active ordered fluids [98,99]. The challenge and interest to the condensed matter physicist is, as always, to ask: how can this long range cooperativity and collective behaviour be understood in terms of the interactions between individuals?

At the modelling level, many of the common features can be captured by the simple rules; follow and avoid [100]. Each individual moves in the average direction of the others it can 'see', while at the same time trying to avoid collisions with any of its neighbours. However, while birds, or people, might enact these rules (semi-)literally, it is more difficult to imagine that the same is true of much simpler organisms like bacteria, for which similar collective behaviour is observed [101–103]. Although it is known that bacteria can detect and respond to chemical gradients [104–106] and to obstacles in the fluid [107] it seems clear that the level at which they do this is different to that of a bird, or wildebeast. Such a simple organism should derive its response from simple principles. In this regard, recent research has focused on the role played by hydrodynamic interactions in determining the collective motion of bacteria and other simple



Figure 5.1: Examples of microscale locomotion. (a) *Echerichia coli* bacteria swim by rotating helical flagella, from Reference [128]. (b) Water striders move at the surface of a liquid, exploiting surface tension for their motility, from Reference [124]. (c) Collective swarming in a sessile drop of *Bacillus subtilis*, from Reference [103].

self-propelled particles [99, 108–113].

The means by which living things achieve their locomotion vary greatly depending on their environment (Fig. 5.1). While fish are able to swim by waving their tails much like a rudder, such motions are not successful for very small organisms such as bacteria, or sperm [114,115]. This is because the inertial forces which provide the thrust for a fish are vastly overwhelmed by the effects of viscous stresses in the case of bacteria. When an organism of characteristic length L moves at speed U through a fluid of viscosity μ and density ρ , the ratio of the inertial and viscous forces defines the Reynolds number for the flow

$$\operatorname{Re} = \frac{\varrho UL}{\mu} \,. \tag{5.1}$$

For most fish the Reynolds number is of the order of several thousands and inertia is important, while for micron sized bacteria it is more typically $\sim 10^{-5}$ and inertia is unimportant. For such small Reynolds numbers the inertial terms in the Navier-Stokes equations may be neglected and the fluid motion adequately described by the Stokes equations

$$\mu \nabla^2 \mathbf{u} - \nabla p = 0 , \qquad (5.2)$$

$$\nabla \cdot \mathbf{u} = 0 , \qquad (5.3)$$

where p is the pressure in the fluid. With the explicit time dependence of the inertial terms gone, the flow is specified throughout the fluid by the instantaneous motion at the boundaries.

The distinctive properties of viscous fluid motion were beautifully illustrated by Sir Geoffrey

Taylor in his educational film "Low Reynolds number flows" [116]. In one experiment a small amount of dye was added to a viscous fluid contained between two coaxial cylinders. When the outer cylinder was rotated by four turns in the clockwise sense, the dye was smeared through the fluid. However, upon counter-rotating the cylinder by four turns in the anticlockwise sense, the blob of dye was exactly recovered as it had been initially. This, now famous, experiment demonstrates the reversible nature of low Reynolds number flows. Taylor went on to vividly demonstrate the consequences of this reversibility for the propulsion of microscopic organisms by showing that a simple model swimmer utilising a waving rudder is unable to swim in a viscous liquid.

The fact that reciprocal motions do not lead to swimming at low Reynolds number was popularised by Purcell [117] and is commonly referred to as the Scallop theorem. Purcell emphasised further that the speed with which the motion is performed is unimportant. Waving the rudder rapidly in one direction and very slowly in the other does not alter the result: at the end of a complete stroke the model returns exactly to its initial position. This is also true for things that actually swim. The distance that a swimmer moves in one complete swimming stroke is unchanged if the swimmer performs the same stroke twice as fast. Only the sequence of shapes matters [118–120].

Since Taylor's seminal work [114,115] there has been steady progress in our understanding of the propulsion of individual swimmers [119,121–125] and in developing optimal swimming strategies [120,126]. The importance of rotational motions was cemented by Berg and Anderson who demonstrated that many bacteria, such as *Escherichia coli*, swim by the rotation of flagella [127, 128]. The propulsive hydrodynamics of rotating helical filaments was described in detail by Lighthill in his 1975 John von Neumann Lecture [121] and later extended by Higdon [129, 130]. Within the past decade the theory of the swimming of elastic filaments has been developed [123, 131] and applied to describe periodic chirality reversals in bacterial flagella [132, 133] and the entire cell bodies of *Spiroplasma* [134, 135].

Considerable insight in recent years has been gained by the analysis of simple models that capture the essence of swimming whilst remaining analytically tractable, following the examples laid down by Taylor [115] and Purcell [117]. This has included both a more detailed analysis of the models proposed by Taylor and Purcell [136–139] and the proposal of new models based on a small number of linked spheres [140–145] or phoretic effects generated by chemical reactions at the swimmer's surface [146, 147]. Initial experiments are beginning to show the viability of some of these simple models as the basis for artificial microswimmers that may be developed for drug delivery or manipulating payloads in microchannels [148–150].

However, whilst our understanding of the motility of single organisms has developed significantly, much less is known about the detailed hydrodynamics of swimmers; the time averaged flow fields that they generate and the interactions that occur between individuals. These interactions may be expected to be substantial because of the long range 1/r nature of the fluid flow generated by point forces at low Reynolds number. They have been shown to be important in the gyrotactic focusing of bottom heavy algal cells [151], band formation in magnetotactic bacteria [152, 153] and in many aspects of bacterial behaviour near surfaces [154–157].

The flow field generated by a swimmer is often described as being dipolar at large distances [101, 109, 110]. This is because swimmers are *self*-motile and are not dragged through the fluid by external influences, so that, at every instant, they must experience zero resultant force [115, 121]. A multipole description of the flow produced by a swimmer will thus have as its most slowly decaying part a dipolar term, varying with distance as $1/r^2$. The dipolar nature of the far field flow generated by a swimmer seems to be so widely accepted that it is rarely calculated explicitly. A noteable exception is a detailed discussion by Lighthill of the flow field generated by a rotating helical filament [158] in which he demonstrated that the combination of finite length and zero resultant torque on the helix was sufficient to imply that the far field flow decays more rapidly than $1/r^2$. Moreover, in the case of *Spirochetes*, Lighthill showed that

"its three-dimensional flow field remains spatially concentrated - without any algebraically decaying terms in the far field - even when the helix is idealised as one of unbounded extent" [158].

Even in the case when the far field flow does possess a dipolar contribution, it is unclear exactly how far from the swimmer one needs to be before this dipolar character is evidenced. Nor is there presently any information on how this distance depends on the type of swimmer, or the detailed properties of its swimming stroke. A naive expectation might be that the magnitude of the far field flow should scale with the slenderness of the swimmer and the amplitude of the swimming stroke in the same way as the swimming speed, which is in general linearly and quadratically, respectively [114, 119], and that this should apply to all terms in a multipole expansion. The transition between near and far field fluid flow would then occur at a distance determined by purely numerical factors, which might even be generic across many different types of swimmer. We shall show in Chapter 6 that these naive expectations are not borne out for a particular model of linked-sphere swimmers.

Hydrodynamic interactions between swimmers result in each individual being both advected and rotated by the fluid flow generated by the others. It might be thought, therefore, that the interactions will assume the same dipolar far field form as is expected of the fluid flow, an approximation that is often taken as the basis for the development of continuum models of swimmers [99, 108, 109]. However, given that the far field form of the flow is not necessarily dipolar we should perhaps be more cautious in assuming any particular behaviour for the hydrodynamic interactions.

Another common approach is to treat the swimmer only in an effective sense, imagined in its simplest form as a 'body' and a 'thruster' [110–113]. In doing so, these approaches do not fully resolve the details of the swimmer and make no attempt to describe its swimming stroke. Thus there is no sense in which a distinction between transverse and longitudinal modes of propulsion can be made. The shape of the swimmer is also neglected and yet our experience with liquid crystals leads us to expect that long, thin, rod-like swimmers, like that introduced by Purcell [117], will have different collective behaviour than flat, disc-like swimmers, such as the one described by Taylor [115].

More faithful descriptions of the hydrodynamic interactions between two swimmers have been studied using a variety of theoretical models [159–168]. Some of the earliest studies made use of flagella driven micromachines [159, 160] and looked at the propulsive advantage, or disadvantage, of side-by-side and tandem swimming. The interactions between two fixed, parallel, rotating helices were investigated [161], revealing that they do not phase synchronise when driven at constant torque. Ishikawa and co-workers [162–164] have studied in detail a simple 'squirmer' model, matching far field asymptotics onto near field lubrication theory and recently applied their results to the description of collective behaviour in small groups of up to two hundred individuals [165].

Nonetheless there remains a sizeable gap in our understanding of how the specific properties of a given swimmer and its swimming stroke influence the hydrodynamic interactions between them. Even many simple questions remain largely unanswered: are the interactions always attractive, or does this depend on the relative positions of the two swimmers? Does the relative phase of the two swimmers play an important role? How important are the details of the particular swimming stroke in determining the interactions? Or, equivalently, how generic are the hydrodynamic interactions between swimmers?

In the remainder of this thesis we shall develop a description of the hydrodynamics of a simple model of linked-sphere swimmers first introduced by Najafi and Golestanian [140] and attempt to answer some of these questions. In Chapter 6 we shall formulate our hydrodynamic description and analyse in some detail both the time averaged flow field generated by a single swimmer and the form, and origin, of the hydrodynamic interactions between individuals. We apply our results to two specific cases; the long time trajectories of two initially parallel swimmers at the end of Chapter 6 and swimmer scattering in Chapter 7.

Throughout our analysis the presence will be felt of the most overarching facet of low Reynolds number hydrodynamics: the kinematic reversibility of Stokes flows. The importance of this has long been recognised for the motility of an individual [114–117], but its consequences for interactions between swimmers and collective motility are only beginning to come to light. We conclude this Chapter with a description of time reversal symmetry and swimming, leading to an important general result concerning the flow field produced by a swimmer, which we shall call Pooley's corollary.

5.1 T-duality

An important concept in understanding the swimming of microscopic organisms is that the Stokes equations, which govern zero Reynolds number fluid flows, do not possess any intrinsic notion of time. This leads directly to the concept of *T*-duality, which we now describe. Consider any successful swimming stroke A: the time reversed swimming stroke \bar{A} corresponds to an equally successful stroke of the same organism. We refer to this as the *T*-dual swimmer, which may be interpreted as the original swimmer going backwards in time.

As an example, consider a simple model of E. coli in which a spherical cell body is propelled by a helical flagellar bundle. The bacterium has a rotary motor that causes the flagella to rotate [104, 127, 128], thereby generating propulsion with the cell body being pushed through the fluid by the rotation of the flagellar bundle lying behind it. We might say that E. coli is an example of a "pusher". If we view this motion backwards in time, then we would observe that swimming is still generated by the rotation of the flagella, but that instead of pushing the cell body now the flagellar bundle is pulling it from in front. Thus we would call this swimmer a "puller". T-duality is merely the observation that pullers are pushers going backwards in time



Figure 5.2: Schematic illustration of T-duality. (a) A pusher, such as *E. coli*, viewed backwards in time is a puller. (b) Some swimmers, such as *Spirillum volutans*, look the same forwards and backwards in time.

(Fig. 5.2).

Exactly the same comments can be made about *extensile* and *contractile* swimmers. This terminology derives from the nature of the time averaged flow field produced by the swimmer. At large distances the average flow will, in many cases, be well approximated by a dipolar field, decaying with distance as $1/r^2$. If the fluid flow is away from the swimmer along its direction of motion then the swimmer is said to be extensile, while if the fluid flow is towards the swimmer then it is called contractile. T-duality provides a relationship between the two: a contractile swimmer may be interpreted as an extensile swimmer going backwards in time.

A special group of swimmers are distinguished by having swimming strokes that are unchanged when viewed backwards in time. We refer to such swimmers as *self T-dual*. It is worth being precise here to avoid any potential confusion with reciprocal swimming strokes. A reciprocal stroke is one that is time reversal *invariant*. Such motions do not lead to swimming on account of kinematic reversibility [116,117,119,169]. A self T-dual stroke on the other hand, is one that is time reversal *covariant*. Several commonly used simple model swimmers are self T-dual, for example a sinusoidally waving sheet [114], Purcell's three link swimmer [117], the Najafi-Golestanian swimmer [140], the pushmepullyou swimmer [144] and the twirler (rotating doughnut) [115,138,139]. Moreover, the rigid rotation of a helical filament [121] is an example of a self T-dual swimming stroke, so that a number of bacteria such as *Spirillum volutans* and *Spirochetes* belong to this class to a very good approximation.

5.1.1 Pooley's corollary

In the case of self T-dual swimmers we can deduce an immediate corollary about the nature of the fluid flow field that they generate. Since their swimming stroke is unchanged when viewed backwards in time, their flow field must be too. Therefore, at large distances, it can be neither extensile, nor contractile, since these look different under time reversal. Thus we conclude that the far field flow of a self T-dual swimmer is not dipolar, but rather decays more rapidly and at least as fast as $1/r^3$.

We shall see an explicit example of this result when we analyse the hydrodynamics of the Najafi-Golestanian swimmer in Chapter 6, however, in agreement with these general symmetry arguments, Lighthill has already shown that the far field flow produced by a helical filament decays more rapidly than $1/r^2$ [158].

CHAPTER 6

Hydrodynamics of linked sphere model swimmers

6.1 The Najafi-Golestanian swimmer

We now describe a simple model low Reynolds number swimmer that was first introduced by Najafi and Golestanian [140] and which we shall take as the basis for much of our analysis. The swimmer consists of three spheres, each of radius a, aligned colinearly and connected by thin rods of 'natural' length D. The rods are made to extend and contract, e.g., by the imagined action of an internal motor, in a periodic and non-time reversible manner. In the original construction, shown in Fig. 6.1, the swimming stroke was broken up into four separate stages, during which the length of only one of the rods was altered. The spheres are labelled 1, 2 and 3, as indicated in Fig. 6.1, increasing in the direction \mathbf{n} , parallel to its long axis and along the direction in which it swims. The hydrodynamics of the model is greatly simplified if we assume that the three spheres are small and far apart, $a/D \ll 1$, and that the rods connecting them are sufficiently thin for their hydrodynamic effect to be neglected. Such sphere plus 'phantom' linker models have become increasingly popular in recent years [144, 145, 166].

In this Chapter we will study a variant of this model in which the four stage swimming stroke is replaced by sinusoidal oscillations of the arm amplitudes. Specifically, the length of the rear and front arms are taken to be

$$D + \xi^{\mathrm{R}} \sin(\omega t)$$
, and $D + \xi^{\mathrm{F}} \sin(\omega t - \phi)$, (6.1)

respectively. Here, we are allowing for the amplitudes of the two oscillations to be different, a feature of some importance. Reversing the direction of time, one sees that the T-dual swimmer



Figure 6.1: Illustration of the Najafi-Golestanian swimmer. The left box shows the original four stage swimming stroke described in Reference [140]. The right box shows the sinusoidal version that we consider here.

corresponds to an interchange of the amplitudes, $\xi^{R} \leftrightarrow \xi^{F}$, and that the swimmer is self T-dual if the amplitudes are equal. Breaking this symmetry by allowing the two arm amplitudes to be different leads to qualitative changes in the hydrodynamic interactions between swimmers. Throughout this Chapter we employ a short-hand notation

$$\tilde{\xi}^{\mathrm{R}} := \xi^{\mathrm{R}} \sin(\omega t) , \qquad (6.2)$$

$$\tilde{\xi}^{\rm F} := \xi^{\rm F} \sin(\omega t - \phi) , \qquad (6.3)$$

to reduce the length of already lengthy formulae.

6.2 Hydrodynamics of a single swimmer

The linearity of the Stokes equations allows the velocity field generated by any low Reynolds number flow to be written as a linear combination of the forces acting on the fluid

$$\mathbf{u}(\mathbf{x}) = \sum_{r} \mathbf{G}^{r}(\mathbf{x}) \mathbf{f}^{r} , \qquad (6.4)$$

where \mathbf{f}^r is the force acting on sphere r of the swimmer and $\mathbf{G}^r(\mathbf{x})$ is the appropriate Green function for the fluid domain under consideration. The problem in applying Eq. (6.4) in practice is that neither the forces, nor the Green function are known exactly. A common approximation, relevant to a dilute suspension of spherical objects in an unbounded fluid, is to take the Green function to be the Oseen tensor [170]

$$\mathbf{G}^{r}(\mathbf{x}) = \begin{cases} \frac{1}{6\pi\mu a} \mathbf{I} & \text{if } |\mathbf{x} - \mathbf{x}^{r}| = a ,\\ \frac{1}{8\pi\mu} \frac{1}{|\mathbf{x} - \mathbf{x}^{r}|} \left(\mathbf{I} + \frac{(\mathbf{x} - \mathbf{x}^{r}) \otimes (\mathbf{x} - \mathbf{x}^{r})}{|\mathbf{x} - \mathbf{x}^{r}|^{2}} \right) & \text{otherwise }, \end{cases}$$
(6.5)

where μ is the fluid viscosity and a is the sphere radius. We shall adopt this approximation for the remainder of this thesis.

The fluid motion is therefore entirely determined by a knowledge of the forces \mathbf{f}^r with which the swimmer acts on the fluid as a result of its changing shape. These are determined by the combined requirements that each swimmer generates no net force, $\sum_r \mathbf{f}^r = \mathbf{0}$, and that the flow field produced is consistent with the shape changes that it undergoes during its swimming stroke

$$\mathbf{u}(\mathbf{x}^2) - \mathbf{u}(\mathbf{x}^1) =: \mathbf{b}^{\mathrm{R}} = \left(\partial_t \tilde{\xi}^{\mathrm{R}}\right) \mathbf{n} , \qquad (6.6)$$

$$\mathbf{u}(\mathbf{x}^3) - \mathbf{u}(\mathbf{x}^2) =: \mathbf{b}^{\mathrm{F}} = \left(\partial_t \tilde{\xi}^{\mathrm{F}}\right) \mathbf{n} .$$
(6.7)

Inserting Eq. (6.4) for the fluid flow converts these consistency requirements into a relationship between the changing shape of the swimmer and the forces with which it acts on the fluid

$$\left\{\frac{1}{6\pi\mu a}\mathbf{I}\begin{pmatrix} -2 & -1\\ 1 & 2 \end{pmatrix} + \mathbf{G}^{1}(\mathbf{x}^{2})\begin{pmatrix} 2 & 1\\ -1 & 0 \end{pmatrix} + \mathbf{G}^{2}(\mathbf{x}^{3})\begin{pmatrix} 0 & 1\\ -1 & -2 \end{pmatrix} + \mathbf{G}^{1}(\mathbf{x}^{3})\begin{pmatrix} 0 & -1\\ 1 & 0 \end{pmatrix}\right\} \begin{pmatrix} \mathbf{f}^{1}\\ \mathbf{f}^{3} \end{pmatrix} = \begin{pmatrix} \mathbf{b}^{\mathrm{R}}\\ \mathbf{b}^{\mathrm{F}} \end{pmatrix}, \quad (6.8)$$

where we have eliminated \mathbf{f}^2 using the force-free constraint. In our approach the shape of the swimmer is specified as a function of time and the forces are the unknowns, which are determined by inverting Eq. (6.8). The simple linear geometry of the Najafi-Golestanian swimmer allows this inverse to be calculated exactly, however, in anticipation of our subsequent calculations we will not pursue this, and instead give a perturbative inversion, using a/D as a small parameter

$$\begin{pmatrix} \mathbf{f}^{1} \\ \mathbf{f}^{3} \end{pmatrix} = 2\pi\mu a \left\{ \mathbf{b}^{\mathrm{R}} \begin{pmatrix} -2 \\ 1 \end{pmatrix} + \mathbf{b}^{\mathrm{F}} \begin{pmatrix} -1 \\ 2 \end{pmatrix} - 2\pi\mu a \left[\mathbf{G}^{1}(\mathbf{x}^{2}) \left\{ \mathbf{b}^{\mathrm{R}} \begin{pmatrix} 4 \\ 1 \end{pmatrix} + \mathbf{b}^{\mathrm{F}} \begin{pmatrix} -1 \\ 2 \end{pmatrix} \right\} \right.$$
$$+ \mathbf{G}^{2}(\mathbf{x}^{3}) \left\{ \mathbf{b}^{\mathrm{R}} \begin{pmatrix} -2 \\ 1 \end{pmatrix} + \mathbf{b}^{\mathrm{F}} \begin{pmatrix} -1 \\ -4 \end{pmatrix} \right\} + \mathbf{G}^{1}(\mathbf{x}^{3}) \left\{ \mathbf{b}^{\mathrm{R}} \begin{pmatrix} 4 \\ -5 \end{pmatrix} + \mathbf{b}^{\mathrm{F}} \begin{pmatrix} 5 \\ -4 \end{pmatrix} \right\} \right] + o\left([a/D]^{2} \right) \right\}.$$
(6.9)

6.2.1 Translational motion

We now describe the translational motion of the swimmer through the fluid by determining its position as a function of time. Once the position of any one of the spheres is known, the positions of the others follow from the prescibed shape of the swimmer, hence it suffices to keep track of only one of them. We choose to track the position of the centre sphere, although this choice is somewhat arbitrary and any other choice would be just as good.

In the zero Reynolds number limit the velocity of each sphere is required to match instantaneously the local velocity of the fluid surrounding it, so that the position of the sphere evolves according to

$$\frac{\mathrm{d}\mathbf{x}^2}{\mathrm{d}t} = \mathbf{u}(\mathbf{x}^2) , \qquad (6.10)$$
$$= \frac{1}{2} (\mathbf{b}^{\mathrm{R}} - \mathbf{b}^{\mathrm{F}}) + \frac{2\pi\mu a}{2} \Big\{ \mathbf{G}^2(\mathbf{x}^3) [2\mathbf{b}^{\mathrm{R}} + \mathbf{b}^{\mathrm{F}}] \Big\}$$

$$= \mathbf{G}^{1}(\mathbf{x}^{2}) \left[\mathbf{b}^{\mathrm{R}} + 2\mathbf{b}^{\mathrm{F}} \right] + \mathbf{G}^{1}(\mathbf{x}^{3}) \left[\mathbf{b}^{\mathrm{F}} - \mathbf{b}^{\mathrm{R}} \right] \right\} + o\left([a/D]^{2} \right).$$

$$(6.11)$$

Integrating this expression gives the displacement of the centre sphere from its initial position as

$$\delta \mathbf{x}^{2}(t) = \mathbf{n} \int_{0}^{t} dt' \left\{ \frac{1}{3} \partial_{t'} \left(\tilde{\xi}^{\mathrm{R}} - \tilde{\xi}^{\mathrm{F}} \right) + \frac{a}{6} \left[\frac{1}{D + \tilde{\xi}^{\mathrm{F}}} \partial_{t'} \left(2 \tilde{\xi}^{\mathrm{R}} + \tilde{\xi}^{\mathrm{F}} \right) - \frac{1}{D + \tilde{\xi}^{\mathrm{R}}} \partial_{t'} \left(\tilde{\xi}^{\mathrm{R}} + 2 \tilde{\xi}^{\mathrm{F}} \right) + \frac{1}{2D + \tilde{\xi}^{\mathrm{R}} + \tilde{\xi}^{\mathrm{F}}} \partial_{t'} \left(\tilde{\xi}^{\mathrm{F}} - \tilde{\xi}^{\mathrm{R}} \right) \right] + o\left([a/D]^{2} \right) \right\}.$$

$$(6.12)$$

The first term describes an $o(\xi)$ oscillation about its initial position that integrates to zero over a complete swimming stroke. This is precisely the motion the sphere would undergo if the swimmer was placed in vacuum and corresponds to the centre of mass of the swimmer remaining fixed. Since the swimmer is in a fluid, and not in vacuum, this is not the only contribution to its motion. The additional terms at o(a/D), and higher, arise due to hydrodynamic interactions and describe swimming, since they do not integrate to zero over a complete swimming stroke. Performing the integration we find that to leading order the total distance moved after each stroke is

$$\mathcal{T} = \frac{2\pi a}{3} \frac{\xi^{\mathrm{R}} \xi^{\mathrm{F}} \sin(\phi)}{D^{2}} \left\{ \left(D/\xi^{\mathrm{R}} \right)^{2} \left[\left(1 - (\xi^{\mathrm{R}}/D)^{2} \right)^{-1/2} - 1 \right] + \left(D/\xi^{\mathrm{F}} \right)^{2} \left[\left(1 - (\xi^{\mathrm{F}}/D)^{2} \right)^{-1/2} - 1 \right] - \frac{1}{4} (2D/\Xi)^{2} \left[\left(1 - (\Xi/2D)^{2} \right)^{-1/2} - 1 \right] \right\} + o\left([a/D]^{2} \right) ,$$

$$(6.13)$$

where

$$\Xi = \left((\xi^{\rm R})^2 + 2\xi^{\rm R}\xi^{\rm F}\cos(\phi) + (\xi^{\rm F})^2 \right)^{1/2}.$$
 (6.14)

6.2.2 Time averaged flow field

The motion of the swimmer constitutes only one part of its hydrodynamics. At low Reynolds numbers fluid disturbances often extend appreciable distances from their point of origin, exercising considerable influence over other objects in the flow. The net flow field generated by a swimmer over the course of its swimming stroke is one such disturbance, whose influence at large distances we now describe.

The time averaged flow field produced by a Najafi-Golestanian swimmer is given by

$$\bar{\mathbf{u}}(\mathbf{x}) := \frac{1}{T} \int_0^T \mathrm{d}t \ \mathbf{u}(\mathbf{x}) = \frac{1}{T} \int_0^T \mathrm{d}t \ \sum_r \mathbf{G}^r(\mathbf{x}) \mathbf{f}^r \ .$$
(6.15)

Part of the difficulty in evaluating this average flow is that the position of the swimmer changes with time, so that the distance $|\mathbf{x} - \mathbf{x}^r|$ is not a constant. Although the positions of each of the spheres, $\mathbf{x}^r(t)$, are in principle known from Eq. (6.12), the use of this directly does not lead to any simple, or convenient expressions for the average flow. However, in the far field, at distances large compared to the size of the swimmer, the magnitude of the oscillations in $\mathbf{x}^r(t)$ will be small compared to the distance to the point of observation. With this in mind we write

$$\mathbf{x} - \mathbf{x}^{r} = (\mathbf{x} - \mathbf{y}) + (\mathbf{y} - \mathbf{x}^{r}) =: \mathbf{r} - \delta \mathbf{x}^{r} , \qquad (6.16)$$

where \mathbf{y} is a fixed point used to represent the (average) position of the swimmer, and make use of a multipole expansion of the stokeslet. In principle the choice of the point \mathbf{y} is somewhat arbitrary, however, for convenience we shall take it to be the position of the centre sphere at a reference time, t = 0, corresponding to the start of the swimming stroke; $\mathbf{y} = \mathbf{x}^2(t = 0)$. The stokeslet is then expanded as a formal power series in 1/r

$$\mathbf{G}^{r}(\mathbf{x}) =: \frac{1}{8\pi\mu} \sum_{j=0}^{\infty} \left[\mathbf{S}^{(j)}(\mathbf{r}) \right] \left(\delta \mathbf{x}^{r} \right)^{\otimes j}.$$
(6.17)

The expansion coefficients $\mathbf{S}^{(j)}$ are formally defined by Eq. (6.17) from which it may readily

be shown that they are given by

$$S^{(j)}_{\alpha\beta\sigma\ldots\tau}(\mathbf{r}) = \frac{(-)^j}{j!} \,\partial_{\sigma}\ldots\partial_{\tau} \left\{ \frac{1}{r} \left(\delta_{\alpha\beta} + \hat{r}_{\alpha}\hat{r}_{\beta} \right) \right\} \,. \tag{6.18}$$

Inserting this multipole expansion for the stokes lets into Eq. (6.15), the far field average flow becomes

$$\bar{u}_{\alpha}(\mathbf{x}) = \sum_{j=0}^{\infty} S^{(j)}_{\alpha\beta\,\sigma\ldots\tau}(\mathbf{r}) n_{\beta} n_{\sigma}\ldots n_{\tau} \int_{0}^{T} \frac{\mathrm{d}t}{T} \sum_{r} \left(\delta x^{r}\right)^{j} \frac{1}{8\pi\mu} f^{r} , \qquad (6.19)$$

since for the Najafi-Golestanian swimmer both the forces \mathbf{f}^r and the displacements $\delta \mathbf{x}^r$ are parallel to the swimming direction \mathbf{n} . Eq. (6.19) is composed of two factors; the tensorial term $\mathbf{S}^{(j)}\mathbf{n}^{\otimes(j+1)}$ which describes how the flow depends on position relative to the swimmer and how it decays with distance, and an integral which captures how the details of the swimming stroke determine the coefficient of each term in the multipole expansion. We can see immediately that the leading j = 0 term vanishes on account of the total force generated by the swimmer being zero. We shall calculate both of the next two terms; the j = 1 dipolar, and j = 2 quadrupolar, contributions to the far field flow.

Dipolar flow

The most slowly decaying term is the $1/r^2$ dipolar contribution, so at large distances we can expect this to provide an accurate description of the average flow field generated by most swimmers. To calculate the integral in Eq. (6.19) we make use of the useful relations

$$\delta x^{1} = \delta x^{2} - \left(D + \tilde{\xi}^{\mathrm{R}}\right), \quad \text{and} \quad \delta x^{3} = \delta x^{2} + \left(D + \tilde{\xi}^{\mathrm{F}}\right), \tag{6.20}$$

which allow us to write

$$\sum_{r} (\delta x^{r}) f^{r} = (D + \tilde{\xi}^{\mathrm{F}}) f^{3} - (D + \tilde{\xi}^{\mathrm{R}}) f^{1} ,$$

$$= \frac{1}{2} (2D + \tilde{\xi}^{\mathrm{R}} + \tilde{\xi}^{\mathrm{F}}) (f^{3} - f^{1}) + \frac{1}{2} (\tilde{\xi}^{\mathrm{F}} - \tilde{\xi}^{\mathrm{R}}) (f^{3} + f^{1}) .$$
(6.21)

Inserting Eq. (6.9) for the forces we find that the relevant integral for the dipolar flow field is

$$\frac{a}{8T} \int_{0}^{T} dt \left\{ 3 \left(2D + \tilde{\xi}^{\mathrm{R}} + \tilde{\xi}^{\mathrm{F}} + \frac{3a}{2} \right) \partial_{t} \left(\tilde{\xi}^{\mathrm{R}} + \tilde{\xi}^{\mathrm{F}} \right) + \left(\tilde{\xi}^{\mathrm{R}} - \tilde{\xi}^{\mathrm{F}} \right) \partial_{t} \left(\tilde{\xi}^{\mathrm{R}} - \tilde{\xi}^{\mathrm{F}} \right) \\ + \frac{a}{2} \left(\tilde{\xi}^{\mathrm{R}} - \tilde{\xi}^{\mathrm{F}} \right) \left[\frac{1}{D + \tilde{\xi}^{\mathrm{R}}} \partial_{t} \left(2\tilde{\xi}^{\mathrm{R}} + 4\tilde{\xi}^{\mathrm{F}} \right) - \frac{1}{D + \tilde{\xi}^{\mathrm{F}}} \partial_{t} \left(4\tilde{\xi}^{\mathrm{R}} + 2\tilde{\xi}^{\mathrm{F}} \right) - \frac{1}{2D + \tilde{\xi}^{\mathrm{R}} + \tilde{\xi}^{\mathrm{F}}} \partial_{t} \left(\tilde{\xi}^{\mathrm{R}} - \tilde{\xi}^{\mathrm{F}} \right) \right] \right\}.$$

$$(6.22)$$

The first line is a total derivative and thus does not contribute to the final result. The remaining integrals can be done using standard techniques to give

$$\frac{\omega a^{2} \sin(\phi)}{8D} \left\{ 2\xi^{\mathrm{F}} \left(\xi^{\mathrm{R}} - \xi^{\mathrm{F}} \cos(\phi)\right) \left(\frac{D}{\xi^{\mathrm{R}}}\right)^{2} \left[\left(1 - \left(\frac{\xi^{\mathrm{R}}}{D}\right)^{2}\right)^{-1/2} - 1 \right] \right. \\ \left. + \xi^{\mathrm{F}} \left(\xi^{\mathrm{R}} + 2\xi^{\mathrm{F}} \cos(\phi)\right) \left(\frac{D}{\xi^{\mathrm{R}}}\right)^{2} \left[1 - \left(1 - \left(\frac{\xi^{\mathrm{R}}}{D}\right)^{2}\right)^{1/2} \right] \right. \\ \left. - 2\xi^{\mathrm{R}} \left(\xi^{\mathrm{F}} - \xi^{\mathrm{R}} \cos(\phi)\right) \left(\frac{D}{\xi^{\mathrm{F}}}\right)^{2} \left[\left(1 - \left(\frac{\xi^{\mathrm{F}}}{D}\right)^{2}\right)^{-1/2} - 1 \right] \right. \\ \left. - \xi^{\mathrm{R}} \left(\xi^{\mathrm{F}} + 2\xi^{\mathrm{R}} \cos(\phi)\right) \left(\frac{D}{\xi^{\mathrm{F}}}\right)^{2} \left[1 - \left(1 - \left(\frac{\xi^{\mathrm{F}}}{D}\right)^{2}\right)^{1/2} \right] \right. \\ \left. - \frac{\xi^{\mathrm{R}} \xi^{\mathrm{F}} (\xi^{\mathrm{R}} + \xi^{\mathrm{F}}) (\xi^{\mathrm{R}} - \xi^{\mathrm{F}})}{8D^{2}} \left(\frac{2D}{\Xi}\right)^{4} \left[2 - \left(1 - \left(\frac{\Xi}{2D}\right)^{2}\right)^{1/2} - \left(1 - \left(\frac{\Xi}{2D}\right)^{2}\right)^{-1/2} \right] \right\} ,$$

$$\left. \left. \left. \left(\frac{1}{2D} \right)^{2} \right] \right\} \right\}$$

where Ξ is again given by Eq. (6.14). Although this expression is quite lengthy and complicated its most important feature is readily apparent: namely it vanishes identically if $\xi^{R} = \xi^{F}$. This, of course, was to be expected. The Najafi-Golestanian swimmer with equal arm amplitudes is self T-dual and thus, by Pooley's corollary (§ 5.1.1), does not have a dipolar contribution to its time averaged flow.

Quadrupolar flow

The quadrupolar term (j = 2) also makes an important contribution to the far field flow of a Najafi-Golestanian swimmer. It decays more rapidly than the dipolar term, varying with distance as $1/r^3$, however its amplitude turns out to be substantially larger so that over a considerable range of intermediate distances from the swimmer, the time averaged flow field may be well approximated as quadrupolar.

To evaluate the integral in Eq. (6.19) we again make use of the relations in Eq. (6.20) to

write

$$\sum_{r} (\delta x^{r})^{2} f^{r} = (f^{3} + f^{1}) \left[\delta x^{2} \left(\tilde{\xi}^{\mathrm{F}} - \tilde{\xi}^{\mathrm{R}} \right) + \left(D + \frac{1}{2} (\tilde{\xi}^{\mathrm{F}} + \tilde{\xi}^{\mathrm{R}}) \right)^{2} + \frac{1}{4} \left(\tilde{\xi}^{\mathrm{F}} - \tilde{\xi}^{\mathrm{R}} \right)^{2} \right] + \left(f^{3} - f^{1} \right) \left(D + \frac{1}{2} (\tilde{\xi}^{\mathrm{F}} + \tilde{\xi}^{\mathrm{R}}) \right) \left(2\delta x^{2} + \tilde{\xi}^{\mathrm{F}} - \tilde{\xi}^{\mathrm{R}} \right) .$$
(6.24)

Inserting the expressions for the forces from Eq. (6.9) and retaining only terms of quadratic order in ξ the integral in Eq. (6.19) becomes

$$\frac{aD}{4T} \int_0^T \mathrm{d}t \left\{ \left[1 + \frac{19a}{8D} \right] \left(\tilde{\xi}^{\mathrm{F}} + \tilde{\xi}^{\mathrm{R}} \right) \partial_t \left(\tilde{\xi}^{\mathrm{F}} - \tilde{\xi}^{\mathrm{R}} \right) + 3 \left[1 + \frac{3a}{4D} \right] \left(2\delta x^2 + \tilde{\xi}^{\mathrm{F}} - \tilde{\xi}^{\mathrm{R}} \right) \partial_t \left(\tilde{\xi}^{\mathrm{F}} + \tilde{\xi}^{\mathrm{R}} \right) \right\}. \quad (6.25)$$

We integrate the second term by parts and use Eq. (6.12) to substitute for $\partial_t \delta x^2$ to find that Eq. (6.25) reduces to

$$\frac{17a^2}{32T} \int_0^T \mathrm{d}t \,\left(\tilde{\xi}^{\mathrm{F}} + \tilde{\xi}^{\mathrm{R}}\right) \partial_t \left(\tilde{\xi}^{\mathrm{F}} - \tilde{\xi}^{\mathrm{R}}\right) = \frac{17\omega a^2 \xi^{\mathrm{R}} \xi^{\mathrm{F}} \sin(\phi)}{32} \,. \tag{6.26}$$

Far field flow

These two contributions, the dipolar and quadrupolar terms, provide a good description of the far field properties of the average flow generated by the swimmer. Combining Eqs. (6.23) and (6.26) with Eq. (6.19) we find that the average far field flow is

$$\bar{\mathbf{u}}(\mathbf{x}) = \frac{21\omega a^2 \xi^{\mathrm{R}} \xi^{\mathrm{F}} \left((\xi^{\mathrm{R}})^2 - (\xi^{\mathrm{F}})^2 \right) \sin(\phi)}{256D^3 r^2} \left\{ \left[3 \left(\mathbf{n} \cdot \hat{\mathbf{r}} \right)^2 - 1 \right] \hat{\mathbf{r}} \right\} + \frac{17\omega a^2 \xi^{\mathrm{R}} \xi^{\mathrm{F}} \sin(\phi)}{64r^3} \left\{ 3 \left(\mathbf{n} \cdot \hat{\mathbf{r}} \right)^2 - 3 \right] \hat{\mathbf{r}} - \left[3 \left(\mathbf{n} \cdot \hat{\mathbf{r}} \right)^2 - 1 \right] \mathbf{n} \right\} + o(1/r^4) ,$$
(6.27)

where we have used the lowest order term in a series expansion in ξ/D for the amplitude of the dipolar term. Comparing the amplitudes of these two contributions at points along the swimming direction, we find that the flow field only becomes dipolar for distances

$$\frac{r}{D} \gtrsim \frac{136D^2}{21(\xi^{\rm R} + \xi^{\rm F})|\xi^{\rm R} - \xi^{\rm F}|} \,. \tag{6.28}$$

This is a remarkable result, which shows that it is only appropriate to view the Najafi-Golestanian swimmer as a simple force-dipole at distances of several tens of body lengths from the swimmer. At closer distances the quadrupolar flow is more significant. However, it should be cautioned that this result is in a sense a worst case scenario. The amplitude of the

dipolar term is required to vanish if either $\xi^{R} = 0$ or $\xi^{F} = 0$, since then the swimming stroke is reciprocal, and also when $\xi^{R} = \xi^{F}$ and the swimmer is self T-dual. This means that the amplitude of the dipolar term must include a factor $\xi^{R}\xi^{F}(\xi^{R} - \xi^{F})$. For sinusoidal strokes with a single frequency, terms of cubic order in the oscillations all integrate to zero, leading to the dipolar flow field scaling as ξ^{4} , as in Eq. (6.27). However, this is not true for other swimming strokes: in particular, for the original, four-stage swimming stroke [140] we have found that the amplitude of the dipolar flow scales as ξ^{3} , and that this dominates the far field time averaged flow for distances [166]

$$\frac{r}{D} \gtrsim \frac{68D}{29|\xi^{\mathrm{R}} - \xi^{\mathrm{F}}|},\tag{6.29}$$

a rather more conservative result than Eq. (6.28).

The time averaged velocity field generated by the Najafi-Golestanian swimmer, using the original four-stage swimming stroke, is shown in Fig. 6.2. It was obtained using a numerical integration of the Oseen tensor equation, subject to the constraint that the swimmer is force and torque-free. Details of this method are given in Reference [145]. Since the numerical technique does not exploit a far field expansion of the stokeslets it enables the flow near the swimmer to be determined as well, as illustrated in the inset. Close to the middle sphere the fluid is clearly seen to move to the left; essentially the swimmer acts like a pump which moves fluid from in front of it to behind it, leading to the swimmer moving to the right. The analytic far field expansion, Eq. (6.27), accurately describes (to within 10%) the numerical flow for distances further than around three swimmer lengths. The dashed circle demarcates the estimated distance, Eq. (6.29), at which the far field flow crosses over from r^{-3} to r^{-2} dipolar scaling.

It is interesting that two separate cases arise for the cross-over to a dipolar far field flow. This is because the dipolar term is antisymmetric under $\xi^{R} \leftrightarrow \xi^{F}$ while the quadrupolar term is symmetric, giving distinct flow fields for the two cases $\xi^{R} < \xi^{F}$ (Fig. 6.2(a)) and $\xi^{R} > \xi^{F}$ (Fig. 6.2(b)). As we shall show, this distinction results in significantly different long time behaviour of interacting swimmers.



Figure 6.2: The flow field around a swimmer, averaged over one swimming cycle. The direction of swimming is from left to right and the flow is axisymmetric about the swimmer. The magnitude of velocities are plotted on a logarithmic scale to enable both the near sphere (inset) and far field behaviour to be observed simultaneously. The parameters used were a = 0.1D, (a) $\xi^{\rm R} = 0.2D, \xi^{\rm F} = 0.4D$ for contractile swimmers, and (b) $\xi^{\rm R} = 0.4D, \xi^{\rm F} = 0.2D$ for extensile swimmers.

6.3 Swimming with friends

We now turn our attention to the hydrodynamics of more than one swimmer. That is, we wish to ask what will be the influence on one swimmer of the presence of other swimmers nearby in the fluid.

Our approach to calculating the hydrodynamics of a group of swimmers parallels our analysis of a single swimmer. In particular, the starting point is again the statement that linearity of the Stokes equations allows the fluid flow to be written as a linear combination of the forces acting on the fluid

$$\mathbf{u}(\mathbf{x}) = \sum_{A} \sum_{r} \mathbf{G}_{A}^{r}(\mathbf{x}) \mathbf{f}_{A}^{r} .$$
(6.30)

Here the subscript A labels the individual swimmers and the other notation is the same as in Eq. (6.4). As for the single swimmer, we will consider that this expression provides a complete solution if the forces \mathbf{f}_A^r can be determined in terms of the prescribed swimming stroke. The constraint that each swimmer is force-free still applies, $\sum_r \mathbf{f}_A^r = \mathbf{0} \forall A$, however the consistency relations on the fluid flow need to be modified to read (for each swimmer)

$$\mathbf{u}(\mathbf{x}_{A}^{2}) - \mathbf{u}(\mathbf{x}_{A}^{1}) =: \mathbf{b}_{A}^{\mathrm{R}} = \left(\partial_{t} \tilde{\xi}_{A}^{\mathrm{R}}\right) \mathbf{n}^{A} + \left[\Omega^{A}, \left(D + \tilde{\xi}_{A}^{\mathrm{R}}\right) \mathbf{n}^{A}\right],$$
(6.31)

$$\mathbf{u}(\mathbf{x}_{A}^{3}) - \mathbf{u}(\mathbf{x}_{A}^{2}) =: \mathbf{b}_{A}^{\mathrm{F}} = \left(\partial_{t}\tilde{\xi}_{A}^{\mathrm{F}}\right)\mathbf{n}^{A} + \left[\Omega^{A}, \left(D + \tilde{\xi}_{A}^{\mathrm{F}}\right)\mathbf{n}^{A}\right], \qquad (6.32)$$

in order to account for the rotational motion of the swimmer. Here we employ an unconventional bracket notation, [,], to denote the vector cross product. The angular velocity Ω^A of each swimmer is determined by imposing the further constraint that its motion impart no net torque to the fluid

$$\sum_{r} \left[\mathbf{x}_{A}^{r} - \mathbf{y} \,, \, \mathbf{f}_{A}^{r} \right] = 0 \;. \tag{6.33}$$

The point \mathbf{y} about which the torque is measured is completely arbitrary because of the force-free constraint. This remains true if the swimmer experiences a net external torque, but remains force-free. However, if it is acted upon by an external force then \mathbf{y} should be taken to be the centre of mass.

As before, we use Eqs. (6.30) to (6.32) to set up a system of linear equations relating the independent forces (\mathbf{f}_A^2 being eliminated via the force-free constraint) to the specified changing

shape of the swimmer. Schematically, these may be written as a matrix equation

$$\sum_{B} \mathcal{G}_{AB} \mathcal{F}_{B} = \mathcal{B}_{A} , \qquad (6.34)$$

where

$$\mathcal{B}_A \coloneqq \begin{pmatrix} \mathbf{b}_A^1 \\ \mathbf{b}_A^2 \end{pmatrix}, \qquad \mathcal{F}_A \coloneqq \begin{pmatrix} \mathbf{f}_A^1 \\ \mathbf{f}_A^3 \end{pmatrix}, \qquad (6.35)$$

$$\mathcal{G}_{AB} := \begin{pmatrix} [\mathbf{G}_B^{12}(\mathbf{x}_A^2) - \mathbf{G}_B^{12}(\mathbf{x}_A^1)] & [\mathbf{G}_B^{32}(\mathbf{x}_A^2) - \mathbf{G}_B^{32}(\mathbf{x}_A^1)] \\ [\mathbf{G}_B^{12}(\mathbf{x}_A^3) - \mathbf{G}_B^{12}(\mathbf{x}_A^2)] & [\mathbf{G}_B^{32}(\mathbf{x}_A^3) - \mathbf{G}_B^{32}(\mathbf{x}_A^2)] \end{pmatrix},$$
(6.36)

and $\mathbf{G}_{B}^{rs}(\mathbf{x}) = \mathbf{G}_{B}^{r}(\mathbf{x}) - \mathbf{G}_{B}^{s}(\mathbf{x})$. The forces will be known if the matrix \mathcal{G}_{AB} can be inverted. To perform the inversion we make use of the fact that the swimmers are in a dilute suspension so that the interactions are all in the weak, far field regime. Consequently the elements \mathcal{G}_{AA} are much larger than \mathcal{G}_{AB} , $A \neq B$, which may be exploited in writing

$$\mathcal{F}_A = \mathcal{G}_{AA}^{-1} \mathcal{B}_A - \sum_{B \neq A} \mathcal{G}_{AA}^{-1} \mathcal{G}_{AB} \mathcal{F}_B .$$
(6.37)

The final solution is then obtained by iteration. The first term represents the forces associated with the swimming of a single isolated organism and are given as before by Eq. (6.9). The second term gives the contribution to the forces due to interactions with all the other swimmers.

6.3.1 Interaction forces: expanding stokeslets

In treating the interactions between swimmers we need to consider objects of the form $\mathbf{G}_B^r(\mathbf{x}_A^s)$, stokeslets associated with spheres comprising swimmer B evaluated at the location of the spheres of swimmer A. In the case of a dilute assembly of swimmers considered here it may be assumed that the separation between swimmers is large compared to the size of any given individual organism. Then, generalising Eq. (6.16), we introduce the decomposition

$$\mathbf{x}_{B}^{r} - \mathbf{x}_{A}^{s} = \left(\mathbf{x}_{B}^{2}(0) - \mathbf{x}_{A}^{2}(0)\right) + \left(\mathbf{x}_{B}^{r} - \mathbf{x}_{B}^{2}(0)\right) - \left(\mathbf{x}_{A}^{s} - \mathbf{x}_{A}^{2}(0)\right),$$

=: $\mathbf{r}_{BA} + \delta \mathbf{x}_{B}^{r} - \delta \mathbf{x}_{A}^{s}$, (6.38)

and perform a multipole expansion of the stokeslets

$$\mathbf{G}_{B}^{r}(\mathbf{x}_{A}^{s}) =: \frac{1}{8\pi\mu} \sum_{j=0}^{\infty} \sum_{k=0}^{j} \left[\mathbf{S}^{(j,k)}(\mathbf{r}_{BA}) \right] \left(\delta \mathbf{x}_{A}^{s} \right)^{\otimes k} \left(\delta \mathbf{x}_{B}^{r} \right)^{\otimes (j-k)} .$$
(6.39)

This decomposition affords a convenient splitting of the interaction into an essentially geometric piece, $\mathbf{S}^{(j,k)}(\mathbf{r}_{BA})$, dependent only on the relative position of the swimmers and terms dependent on the details of the swimming motions. Again, the expansion coefficients $\mathbf{S}^{(j,k)}$ are formally defined by Eq. (6.39) from which it may be shown that they are given by

$$S^{(j,k)}_{\alpha\beta\sigma\ldots\tau}(\mathbf{r}) = \frac{(-)^k}{k!(j-k)!}\partial_{\sigma}\ldots\partial_{\tau}\left\{\frac{1}{r}\left(\delta_{\alpha\beta} + \hat{r}_{\alpha}\hat{r}_{\beta}\right)\right\}.$$
(6.40)

These tensors are fully symmetric in the j indices $\sigma \dots \tau$ as well as being symmetric in the indices α, β . They scale with the separation between swimmers as $r_{BA}^{-(j+1)}$ so that the terms with the lowest values of j are expected to dominate the far field interactions. With this expansion of the stokeslets the contribution to the forces coming from interactions is given by

$$\begin{pmatrix} (f^{\text{INT}})_A^1 \\ (f^{\text{INT}})_A^3 \end{pmatrix}_{\alpha} = \frac{a}{4} \sum_{B \neq A} \sum_{j=2}^{\infty} \sum_{k=1}^{j-1} S^{(j,k)}_{\alpha\beta \sigma \dots \tau \nu \dots \rho}(\mathbf{r}_{BA}) \\ \begin{pmatrix} -2(\delta x_A^1)_{\sigma \dots \tau}^{\otimes k} + (\delta x_A^2)_{\sigma \dots \tau}^{\otimes k} + (\delta x_A^3)_{\sigma \dots \tau}^{\otimes k} \\ (\delta x_A^1)_{\sigma \dots \tau}^{\otimes k} + (\delta x_A^2)_{\sigma \dots \tau}^{\otimes k} - 2(\delta x_A^3)_{\sigma \dots \tau}^{\otimes k} \end{pmatrix} \sum_{r} (\delta x_B^r)_{\nu \dots \rho}^{\otimes (j-k)} (f_B^r)_{\beta} .$$

$$(6.41)$$

6.3.2 Swimmer rotation

Having determined both the single swimmer, Eq. (6.9), and interaction, Eq. (6.41), contributions to the forces, the sole remaining unknown is the angular velocity of the swimmer, Ω^A . This is obtained through imposing the constraint that the swimmer is torque-free, Eq. (6.33), which we write as

$$0 = \sum_{r} \left[\left(\mathbf{x}_{A}^{r} - \mathbf{y} \right), \mathbf{f}_{A}^{r} \right],$$

= $\left[\left(\mathbf{x}_{A}^{3} - \mathbf{x}_{A}^{2} \right), \mathbf{f}_{A}^{3} \right] - \left[\left(\mathbf{x}_{A}^{2} - \mathbf{x}_{A}^{1} \right), \mathbf{f}_{A}^{1} \right],$
= $\left(D + \frac{1}{2} \left(\tilde{\xi}_{A}^{\mathrm{R}} + \tilde{\xi}_{A}^{\mathrm{F}} \right) \right) \left[\mathbf{n}^{A}, \left(\mathbf{f}_{A}^{3} - \mathbf{f}_{A}^{1} \right) \right] + \frac{1}{2} \left(\tilde{\xi}_{A}^{\mathrm{F}} - \tilde{\xi}_{A}^{\mathrm{R}} \right) \left[\mathbf{n}^{A}, \left(\mathbf{f}_{A}^{3} + \mathbf{f}_{A}^{1} \right) \right].$ (6.42)

It only proves necessary to balance the o(1) part of the single swimmer forces, Eq. (6.9), against the contribution due to interactions, Eq. (6.41). There is no fundamental obstacle to retaining the higher order terms from Eq. (6.9), however, since these lead to a substantial increase in the length of formulae and play no essential role, they will be omitted in what follows. After some straightforward manipulations the torque balance equation becomes

$$\epsilon_{\alpha\beta\gamma} \left\{ P \ n_{\beta}^{A} n_{\delta}^{A} \left[\Omega^{A}, \right]_{\gamma\delta} - \frac{a}{4} \sum_{B \neq A} \sum_{j=2}^{\infty} \sum_{k=1}^{j-1} S^{(j,k)}_{\gamma\lambda \sigma \dots \tau \nu \dots \rho}(\mathbf{r}_{BA}) \ n_{\beta}^{A} A^{k}_{\sigma \dots \tau} \ C^{j-k}_{\lambda \nu \dots \rho} \right\} = 0 , \qquad (6.43)$$

where we have defined

$$P := 6 \left(D + \frac{1}{2} (\tilde{\xi}_A^{\rm R} + \tilde{\xi}_A^{\rm F}) \right)^2 + \frac{1}{2} \left(\tilde{\xi}_A^{\rm R} - \tilde{\xi}_A^{\rm F} \right)^2, \qquad (6.44)$$

$$\begin{aligned}
A^{k}_{\sigma\ldots\tau} &\coloneqq 3D\Big((\delta x^{3}_{A})^{\otimes k}_{\sigma\ldots\tau} - (\delta x^{1}_{A})^{\otimes k}_{\sigma\ldots\tau}\Big) + \tilde{\xi}^{\mathrm{R}}_{A}\Big(-2(\delta x^{1}_{A})^{\otimes k}_{\sigma\ldots\tau} + (\delta x^{2}_{A})^{\otimes k}_{\sigma\ldots\tau} + (\delta x^{3}_{A})^{\otimes k}_{\sigma\ldots\tau}\Big) \\
&\quad + \tilde{\xi}^{\mathrm{F}}_{A}\Big(-(\delta x^{1}_{A})^{\otimes k}_{\sigma\ldots\tau} - (\delta x^{2}_{A})^{\otimes k}_{\sigma\ldots\tau} + 2(\delta x^{3}_{A})^{\otimes k}_{\sigma\ldots\tau}\Big),
\end{aligned}$$
(6.45)

$$C^{j-k}_{\lambda \nu \dots \rho} \coloneqq \left(b^1_B\right)_{\lambda} \left[-2(\delta x^1_B)^{\otimes (j-k)}_{\nu \dots \rho} + (\delta x^2_B)^{\otimes (j-k)}_{\nu \dots \rho} + (\delta x^3_B)^{\otimes (j-k)}_{\nu \dots \rho} \right] + \left(b^2_B\right)_{\lambda} \left[-(\delta x^1_B)^{\otimes (j-k)}_{\nu \dots \rho} - (\delta x^2_B)^{\otimes (j-k)}_{\nu \dots \rho} + 2(\delta x^3_B)^{\otimes (j-k)}_{\nu \dots \rho} \right].$$

$$(6.46)$$

This equation is easily solved to give the angular velocity of each swimmer as

$$[\Omega^{A},]_{\alpha\beta} = \frac{a}{4} \sum_{B \neq A} \sum_{j=2}^{\infty} \sum_{k=1}^{j-1} \left(\delta_{\alpha\gamma} n_{\beta}^{A} - \delta_{\beta\gamma} n_{\alpha}^{A} \right) \left[S_{\gamma\lambda \sigma \dots \tau \nu \dots \rho}^{(j,k)}(\mathbf{r}_{BA}) \right] \left(P^{-1} A_{\sigma \dots \tau}^{k} \right) C_{\lambda \nu \dots \rho}^{j-k} .$$
(6.47)

Our main interest is in the net effect that the interactions have over one complete swimming stroke. Now, the orientation of each swimmer evolves according to the equation

$$\frac{\mathrm{d}n^A_\alpha}{\mathrm{d}t} = [\Omega^A, \mathbf{n}^A]_\alpha \ . \tag{6.48}$$

Technically, this is a set of N_{swimmer} coupled, non-linear, ordinary differential equations. We make no attempt to solve it exactly, being satisfied with a perturbative approach. First we solve for the motion of a single swimmer, which we then substitute into the right hand side of Eq. (6.48). This means the right hand side may be considered as simply a function of t and integrated directly. In this case a single swimmer does not rotate, so that where **n** appears on the right hand side it may be considered just a constant vector. The net change in orientation

over a complete swimming stroke is thus

$$n_{\alpha}^{A}(T) - n_{\alpha}^{A} = \frac{a}{4} \sum_{B \neq A} \sum_{j=2}^{\infty} \sum_{k=1}^{j-1} \left(\delta_{\alpha\beta} - n_{\alpha}^{A} n_{\beta}^{A} \right) \left[S_{\beta\gamma \sigma \dots \tau \nu \dots \rho}^{(j,k)}(\mathbf{r}_{BA}) \right] n_{\gamma}^{B} n_{\sigma}^{A} \dots n_{\tau}^{A} n_{\nu}^{B} \dots n_{\rho}^{B} \left(\mathcal{I}^{(j,k)} \right) ,$$

$$\tag{6.49}$$

where

$$\mathcal{I}^{(j,k)} := \int_0^T \mathrm{d}t \ P^{-1} A^k \ C^{j-k} \ . \tag{6.50}$$

Again we emphasise that this expression decomposes the effect of the interactions into two parts: the tensorial part involving $\mathbf{S}^{(j,k)}$ which captures how the relative positions and orientations of the swimmers influence their interactions, and the integral $\mathcal{I}^{(j,k)}$ which captures the details of the swimming stroke. In fact, the form of these integrals offers an important insight into the nature of hydrodynamic interactions between swimmers. A short calculation reveals that $A^1 = P$ so that all terms with k = 1 in Eq. (6.49) are independent of the swimming motion of swimmer A. We refer to these as *passive* interactions. They correspond to the rotation that would be experienced by an inanimate object, or dead swimmer, drifting passively in the flow field generated by the other swimmers. By contrast, the terms with $k \ge 2$ depend on the swimming motion of both swimmers. We call these *active* interactions. They represent the additional rotation experienced by the swimmers because they are trying to swim *simultaneously* and encode all of the information about the relative phase of the swimmers.

The most slowly decaying active term in the far field is the contribution with j = 3, k = 2, which we now determine. A straightforward calculation gives

$$A^{2} = P \left[2\delta x_{A}^{2} + \frac{7}{6} (\tilde{\xi}_{A}^{\mathrm{F}} - \tilde{\xi}_{A}^{\mathrm{R}}) \right] - \frac{1}{3} (\tilde{\xi}_{A}^{\mathrm{F}} - \tilde{\xi}_{A}^{\mathrm{R}})^{3} , \qquad (6.51)$$

$$C^{1} = 3D \partial_{t} \left(\tilde{\xi}_{B}^{\mathrm{F}} + \tilde{\xi}_{B}^{\mathrm{R}} \right) + \partial_{t} \left((\tilde{\xi}_{B}^{\mathrm{F}})^{2} + \tilde{\xi}_{B}^{\mathrm{F}} \tilde{\xi}_{B}^{\mathrm{R}} + (\tilde{\xi}_{B}^{\mathrm{R}})^{2} \right) + o(a/D) , \qquad (6.52)$$

so that the integral $\mathcal{I}^{(3,2)}$ is given by

$$\mathcal{I}^{(3,2)} = \frac{3\pi D}{2} \Big\{ \xi_A^{\rm R} \big[\xi_B^{\rm R} \sin(\eta_{BA}) + \xi_B^{\rm F} \sin(\eta_{BA} - \phi_B) \big] \\ - \xi_A^{\rm F} \big[\xi_B^{\rm R} \sin(\eta_{BA} + \phi_A) + \xi_B^{\rm F} \sin(\eta_{BA} + \phi_A - \phi_B) \big] \Big\} + o(\xi^4) .$$
(6.53)

Here, η_{BA} is the phase of swimmer *B* relative to swimmer *A*, i.e., if $\tilde{\xi}_A^{\mathrm{R}} = \xi_A^{\mathrm{R}} \sin(\omega t)$ then $\tilde{\xi}_B^{\mathrm{R}} = \xi_B^{\mathrm{R}} \sin(\omega t + \eta_{BA})$ and similarly for the front amplitudes. We remark in passing that this amplitude does *not* vanish for $\phi_A = \phi_B = \pi$ when the two swimmers are reciprocal. Instead we

find $\mathcal{I}^{(3,2)} = (3\pi D/2)(\xi_A^R \xi_B^R - \xi_A^F \xi_B^F) \sin(\eta_{BA})$, so that, provided the swimmers are not in phase or exactly out of phase, they can still interact hydrodynamically, despite each individually performing a reciprocal motion. This insight will be discussed in more detail in Chapter 8.

Combining Eqs. (6.49) and (6.53), we find that the active contributions to the interactions lead to a rotation of the swimmers that in the far field has the asymptotic form

$$\Delta \mathbf{n}_{\text{active}}^{A} \sim \sum_{B \neq A} \frac{-3a\mathcal{I}^{(3,2)}}{8r_{BA}^{4}} \Big[1 + 2(\mathbf{n}^{A} \cdot \mathbf{n}^{B})^{2} - 5(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})^{2} - 5(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{2} - 20(\mathbf{n}^{A} \cdot \mathbf{n}^{B})(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA}) + 35(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})^{2}(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{2} \Big] \Big\{ \hat{\mathbf{r}}_{BA} - (\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})\mathbf{n}^{A} \Big\} .$$

$$(6.54)$$

For the passive interactions, the rotation can be determined using the time averaged flow field, Eq. (6.27), from which we obtain

$$\begin{aligned} \boldsymbol{\Delta} \mathbf{n}_{\text{passive}}^{A} &\sim \sum_{B \neq A} \frac{63\pi a^{2} \boldsymbol{\xi}_{B}^{R} \boldsymbol{\xi}_{B}^{F} \left((\boldsymbol{\xi}_{B}^{R})^{2} - (\boldsymbol{\xi}_{B}^{F})^{2} \right) \sin(\phi_{B})}{128 D^{3} r_{BA}^{3}} \Big[(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA}) + 2(\mathbf{n}^{A} \cdot \mathbf{n}^{B})(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA}) \\ &\quad - 5(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{2} \Big] \Big\{ \hat{\mathbf{r}}_{BA} - (\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})\mathbf{n}^{A} \Big\} \\ &\quad + \sum_{B \neq A} \frac{51\pi a^{2} \boldsymbol{\xi}_{B}^{R} \boldsymbol{\xi}_{B}^{F} \sin(\phi_{B})}{32 r_{BA}^{4}} \Big\{ \Big[3(\mathbf{n}^{A} \cdot \mathbf{n}^{B}) - 15(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA}) \\ &\quad - 15(\mathbf{n}^{A} \cdot \mathbf{n}^{B})(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{2} + 35(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{3} \Big] \Big\{ \hat{\mathbf{r}}_{BA} - (\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})\mathbf{n}^{A} \Big\} \\ &\quad + \Big[(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA}) + 2(\mathbf{n}^{A} \cdot \mathbf{n}^{B})(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA}) - 5(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{2} \Big] \\ &\quad \times \Big\{ \mathbf{n}^{B} - (\mathbf{n}^{A} \cdot \mathbf{n}^{B})\mathbf{n}^{A} \Big\} \Big\} . \end{aligned}$$

The most significant implication of this result is that the rotation generated by the active interactions is substantially larger than that of the passive interactions for all reasonable separations. The passive terms scale as $(a/D)^2$, while the active terms scale as a/D. Thus the former are suppressed by an additional factor of the slenderness of the swimmer, which is always a small number for the Najafi-Golestanian swimmer. The important consequence of this is that the relative phase, which enters only into the active terms, plays an important role in the hydrodynamic interactions of microswimmers. This feature, which is central to our work, is not captured by coarse-grained models of interacting swimmers that do not faithfully account for the details of the swimming stroke.

6.3.3 Swimmer advection

In addition to a rotation of their direction of motion, the interactions also give rise to an advection of each swimmer in the flow field produced by the others. This advection enters into the determination of the translational motion of each swimmer described in § 6.2.1. Eq. (6.10) still applies, although with the fluid velocity given by Eq. (6.30)

$$\frac{\mathrm{d}\mathbf{x}_A^2}{\mathrm{d}t} = \mathbf{u}(\mathbf{x}_A^2) = \sum_s \mathbf{G}_A^s(\mathbf{x}_A^2)\mathbf{f}_A^s + \sum_{B \neq A} \sum_r \mathbf{G}_B^r(\mathbf{x}_A^2)\mathbf{f}_B^r \,. \tag{6.56}$$

The advection arises both directly, through the second $\sum_{B \neq A}$ term in Eq. (6.56), and indirectly, from the fact that the forces \mathbf{f}_A^s differ from their values for a single isolated swimmer on account of the interactions. When these are combined we find that the advective contribution to $\mathbf{u}(\mathbf{x}_A^2)$ is, to leading order, given by

$$\frac{a}{12} \sum_{B \neq A} \sum_{j=1}^{\infty} \sum_{k=0}^{j-1} \left[\mathbf{S}^{(j,k)}(\mathbf{r}_{BA}) \right] \left((\delta \mathbf{x}_A^1)^{\otimes k} + (\delta \mathbf{x}_A^2)^{\otimes k} + (\delta \mathbf{x}_A^3)^{\otimes k} \right) \mathbf{C}^{j-k} .$$
(6.57)

The equation for the translational motion, Eq. (6.56), is also a set of coupled, non-linear, ordinary differential equations and hence we employ the same perturbative approach as for the rotation, obtaining

$$[x_{A}^{2}]_{\alpha}(T) - [x_{A}^{2}]_{\alpha} = \mathcal{T}n_{\alpha}^{A} + \frac{a}{12} \sum_{B \neq A} \sum_{j=1}^{\infty} \sum_{k=0}^{j-1} \left[S_{\alpha\beta \ \sigma \dots \tau \ \nu \dots \rho}^{(j,k)}(\mathbf{r}_{BA}) \right] n_{\beta}^{B} n_{\sigma}^{A} \dots n_{\tau}^{A} n_{\nu}^{B} \dots n_{\rho}^{B} \left(\mathcal{J}^{(j,k)} \right) ,$$
(6.58)

where \mathcal{T} is the single swimmer translational motion given by Eq. (6.13) and

$$\mathcal{J}^{(j,k)} := \int_0^T \mathrm{d}t \, \left(\left(\delta x_A^1 \right)^k + \left(\delta x_A^2 \right)^k + \left(\delta x_A^3 \right)^k \right) C^{j-k} \,. \tag{6.59}$$

Again a distinction can be made between passive (k = 0) terms and active $(k \ge 1)$ terms. As for the rotational interaction, the passive advection results from the same calculation that gives the time averaged flow field, Eq. (6.27), and yields the result

$$\begin{aligned} \mathbf{\Delta} \mathbf{x}_{A \text{ passive}}^{2} &\sim \sum_{B \neq A} \frac{21\pi a^{2} \xi_{B}^{\mathrm{R}} \xi_{B}^{\mathrm{F}} \left((\xi_{B}^{\mathrm{R}})^{2} - (\xi_{B}^{\mathrm{F}})^{2} \right) \sin(\phi_{B})}{128 D^{3} r_{BA}^{3}} \left[1 - 3 (\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{2} \right] \hat{\mathbf{r}}_{BA} \\ &+ \sum_{B \neq A} \frac{17\pi a^{2} \xi_{B}^{\mathrm{R}} \xi_{B}^{\mathrm{F}} \sin(\phi_{B})}{32 r_{BA}^{3}} \left\{ 3 (\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA}) \left[5 (\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{2} - 3 \right] \hat{\mathbf{r}}_{BA} - \left[3 (\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{2} - 1 \right] \mathbf{n}^{B} \right\}. \end{aligned}$$

$$(6.60)$$

In addition to this passive advection there will be an active contribution, which we expect to have a substantially larger amplitude, scaling as a/D instead of $(a/D)^2$. Unlike the rotational interaction, this does not come from the first non-trivial term, i.e., j = 2, k = 1. This is because the sum of displacements $\delta x_A^1 + \delta x_A^2 + \delta x_A^3$ is itself o(a/D), since, as discussed following Eq. (6.12), this quantity represents the centre of mass motion of the swimmer, which is only non-zero because of hydrodynamic interactions between the spheres. Thus the leading active interaction comes from the term j = 3, k = 2, for which we find

$$\mathcal{J}^{(3,2)} = -6\pi D^2 \Big\{ \xi_A^{\rm R} [\xi_B^{\rm R} \sin(\eta_{BA}) + \xi_B^{\rm F} \sin(\eta_{BA} - \phi_B)] \\ + \xi_A^{\rm F} [\xi_B^{\rm R} \sin(\eta_{BA} + \phi_A) + \xi_B^{\rm F} \sin(\eta_{BA} + \phi_A - \phi_B)] \Big\} ,$$
(6.61)

and the active advection is then given by

$$\begin{aligned} \mathbf{\Delta}\mathbf{x}_{A \text{ active}}^{2} &\sim \sum_{B \neq A} \frac{-a\mathcal{J}^{(3,2)}}{8r_{BA}^{4}} \bigg\{ \bigg[1 + 2(\mathbf{n}^{A} \cdot \mathbf{n}^{B})^{2} - 5(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})^{2} - 5(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{2} \\ &- 20(\mathbf{n}^{A} \cdot \mathbf{n}^{B})(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA}) + 35(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})^{2}(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{2} \bigg] \hat{\mathbf{r}}_{BA} \\ &+ 2\bigg[(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA}) + 2(\mathbf{n}^{A} \cdot \mathbf{n}^{B})(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA}) - 5(\mathbf{n}^{A} \cdot \hat{\mathbf{r}}_{BA})(\mathbf{n}^{B} \cdot \hat{\mathbf{r}}_{BA})^{2} \bigg] \mathbf{n}^{A} \bigg\} . \end{aligned}$$

6.3.4 An improved near field calculation

A drawback of the approach we have just described is that it is based on a far field analysis, where the separation between swimmers is large compared to the size of an individual, $D/r \ll 1$. Since the Oseen tensor approach allows us to determine the hydrodynamic interactions between two spheres of the *same* swimmer when they are a distance D apart (for the determination of the single swimmer motion), it should also allow us to determine the interactions between two spheres of *different* swimmers when the separation between them is also o(D). Clearly this cannot be done on the basis of a multipole expansion in powers of D/r, since for r = o(D) this will be, at best, very slowly convergent. At these close separations we can replace Eq. (6.38) for the relative position of the two spheres with

$$\mathbf{x}_B^r - \mathbf{x}_A^s = \left(\mathbf{x}_B^r(0) - \mathbf{x}_A^s(0)\right) + \left(\mathbf{x}_B^r - \mathbf{x}_B^r(0)\right) - \left(\mathbf{x}_A^s - \mathbf{x}_A^s(0)\right),$$

$$=: \mathbf{r}_{BA}^{(r,s)} + \delta \mathbf{x}_B^r - \delta \mathbf{x}_A^s.$$
(6.63)

If we consider that the deviation of each sphere from its "mean" position is small compared to the separation between them, $\xi/D \ll 1$, then we can still make use of an expansion of stokeslets to determine the interactions in the same way as before. The price that we pay for this is that the tensors $\mathbf{S}^{(j,k)}$ appearing in Eq. (6.39) are no longer independent of the pair of spheres (r, s), so that each pair needs to be considered individually. This leads to a substantial increase in the length of our formulae. For example, Eq. (6.47) becomes

$$\begin{split} & \left[\Omega^{A}, \right]_{\alpha\beta} = \frac{a}{4}P^{-1} \left(\delta_{\alpha\gamma}n_{\beta}^{A} - \delta_{\beta\gamma}n_{\alpha}^{A}\right) \sum_{B \neq A} \sum_{j=0}^{\infty} \sum_{k=0}^{j} n_{\lambda}^{B} n_{\sigma}^{A} \dots n_{\tau}^{A} n_{\nu}^{B} \dots n_{\rho}^{B} \\ & \left\{ \left[S_{\gamma\lambda\sigma,\dots\tau\nu\dots\rho}^{(j,k)}(\mathbf{r}_{BA}^{(1,1)}) \right] \left(\delta x_{A}^{1}\right)^{k} \left(\delta x_{B}^{1}\right)^{j-k} \left[\left(3D + 2\xi_{A}^{R} + \xi_{A}^{F}\right) \partial_{t} \left(2\xi_{B}^{R} + \xi_{B}^{F}\right) \right] \right. \\ & \left. + \left[S_{\gamma\lambda\sigma,\dots\tau\nu\dots\rho}^{(j,k)}(\mathbf{r}_{BA}^{(1,2)}) \right] \left(\delta x_{A}^{2}\right)^{k} \left(\delta x_{B}^{1}\right)^{j-k} \left[\left(\xi_{A}^{F} - \xi_{A}^{R}\right) \partial_{t} \left(2\xi_{B}^{R} + \xi_{B}^{F}\right) \right] \right. \\ & \left. + \left[S_{\gamma\lambda\sigma,\dots\tau\nu\dots\rho}^{(j,k)}(\mathbf{r}_{BA}^{(1,2)}) \right] \left(\delta x_{A}^{3}\right)^{k} \left(\delta x_{B}^{1}\right)^{j-k} \left[\left(3D + 2\xi_{A}^{R} + 2\xi_{A}^{F}\right) \partial_{t} \left(2\xi_{B}^{R} + \xi_{B}^{F}\right) \right] \right. \\ & \left. + \left[S_{\gamma\lambda\sigma,\dots\tau\nu\dots\rho}^{(j,k)}(\mathbf{r}_{BA}^{(2,1)}) \right] \left(\delta x_{A}^{1}\right)^{k} \left(\delta x_{B}^{2}\right)^{j-k} \left[\left(3D + 2\xi_{A}^{R} + \xi_{A}^{F}\right) \partial_{t} \left(\xi_{B}^{F} - \xi_{B}^{R}\right) \right] \right. \\ & \left. + \left[S_{\gamma\lambda\sigma,\dots\tau\nu\dots\rho}^{(j,k)}(\mathbf{r}_{BA}^{(2,2)}) \right] \left(\delta x_{A}^{2}\right)^{k} \left(\delta x_{B}^{2}\right)^{j-k} \left[\left(-3D + \xi_{A}^{R} + 2\xi_{A}^{F}\right) \partial_{t} \left(\xi_{B}^{F} - \xi_{B}^{R}\right) \right] \right. \\ & \left. + \left[S_{\gamma\lambda\sigma,\dots\tau\nu\dots\rho}^{(j,k)}(\mathbf{r}_{BA}^{(3,1)}) \right] \left(\delta x_{A}^{1}\right)^{k} \left(\delta x_{B}^{3}\right)^{j-k} \left[- \left(3D + 2\xi_{A}^{R} + 2\xi_{A}^{F}\right) \partial_{t} \left(\xi_{B}^{R} - \xi_{B}^{R}\right) \right] \right. \\ & \left. + \left[S_{\gamma\lambda\sigma,\dots\tau\nu\dots\rho}^{(j,k)}(\mathbf{r}_{BA}^{(3,1)}) \right] \left(\delta x_{A}^{1}\right)^{k} \left(\delta x_{B}^{3}\right)^{j-k} \left[- \left(3D + 2\xi_{A}^{R} + 2\xi_{A}^{F}\right) \partial_{t} \left(\xi_{B}^{R} + 2\xi_{B}^{F}\right) \right] \right. \\ & \left. + \left[S_{\gamma\lambda\sigma,\dots\tau\nu\dots\rho}^{(j,k)}(\mathbf{r}_{BA}^{(3,1)}) \right] \left(\delta x_{A}^{2}\right)^{k} \left(\delta x_{B}^{3}\right)^{j-k} \left[- \left(3D + 2\xi_{A}^{R} + \xi_{A}^{F}\right) \partial_{t} \left(\xi_{B}^{R} + 2\xi_{B}^{F}\right) \right] \right. \\ & \left. + \left[S_{\gamma\lambda\sigma,\dots\tau\nu\dots\rho}^{(j,k)}(\mathbf{r}_{BA}^{(3,1)}) \right] \left(\delta x_{A}^{2}\right)^{k} \left(\delta x_{B}^{3}\right)^{j-k} \left[- \left(\xi_{A}^{F} - \xi_{A}^{R}\right) \partial_{t} \left(\xi_{B}^{R} + 2\xi_{B}^{F}\right) \right] \right] \right. \\ \\ & \left. + \left[S_{\gamma\lambda\sigma,\dots\tau\nu\dots\rho}^{(j,k)}(\mathbf{r}_{BA}^{(3,1)}) \right] \left(\delta x_{A}^{3}\right)^{k} \left(\delta x_{B}^{3}\right)^{j-k} \left[\left(3D + \xi_{A}^{R} + 2\xi_{A}^{F}\right) \partial_{t} \left(\xi_{B}^{R} + 2\xi_{B}^{F}\right) \right] \right\} .$$

The leading order contribution to the active interaction comes from the j = 0, k = 0 and j = 1, k = 1 terms and thus can be thought of as a collection of stokeslets, $\mathbf{S}^{(0,0)}$, and stokes doublets, $\mathbf{S}^{(1,1)}$ [171]. The integrals over a complete swimming stroke of these terms are straightforward, but the number of them makes their evaluation tedious and we shall not quote the results here. The advection can be treated in a similar fashion, but again the results will not be quoted explicitly due to the length of the final expressions.

Comparison between near and far field approaches

A comparison of the results of the near and far field calculations is presented in Fig. 6.3 for two parallel, coplanar swimmers. First, in Fig. 6.3(a) we show how the rotation angle $\Delta\theta$ depends on the separation between the swimmers in the near field calculation. This is found to asymptote onto the predicted far field scaling of r_{BA}^{-4} for distances of greater than about 10D, but to become less singular at closer distances. In Figs. 6.3(b) and (c) we show how $\Delta\theta$ varies as one swimmer is moved in a circle around the other, keeping a fixed separation between them. At a separation of 5D (Fig. 6.3(b)) there are noticeable differences between the near and far field calculations, although the qualitative features are the same. These discrepancies are substantially reduced when the separation is increased to 10D (Fig. 6.3(c)), indicating that the far field description is sufficient.

The simulation parameters used for this comparison were a = 0.05D, $\xi^{R} = \xi^{F} = 0.2D$, $\phi_{A} = \phi_{B} = \pi/2$ and $\eta_{BA} = 0$.

6.4 Trajectories of two parallel swimmers

We conclude this Chapter by applying the results of § 6.3 to a simple problem involving the hydrodynamic interactions between just two swimmers. The two swimmers are initially oriented along parallel directions and we consider how their subsequent long time trajectories change as one swimmer is moved in a circle around the other one, keeping them a fixed separation of 5D apart.

We consider first self T-dual swimmers ($\xi^{R} = \xi^{F}$) swimming in phase, before looking at how the behaviour changes when these conditions are relaxed. The results are summarised in Fig. 6.4(a). Swimmer A initially lies at the origin (as shown in the figure) and swimmer B is aligned parallel and displaced to the corresponding point in the diagram. The figure is divided into different regions, labelled A, R, O, and P, corresponding to the four different types of long term behaviour that were observed; attractive, repulsive, oscillatory and parallel trajectories.

Typical trajectories for each of these types of behaviour are also shown in Fig. 6.4(i-iv). For accuracy, the trajectories were obtained numerically [145], although the results which follow from iterating the analytic formulae, Eqs. (6.54) and (6.62), are in close agreement. All scales are in units of D, the swimmer arm length. The parameters chosen were a = 0.1D and $\xi = 0.3D$, giving a displacement per cycle of $\mathcal{T} \approx (7\pi/12)a\xi^2/D^2 = 0.016D$. Thus, since the



Figure 6.3: Comparison of the near and far field calculations of the hydrodynamics of Najafi-Golestanian swimmers. (a) The dependence of the rotation angle on separation for two parallel swimmers, measured in the direction perpendicular to their orientation. (b) and (c) Variation of the rotation angle with the relative position of the two swimmers at a fixed separation of (b) $r_{BA} = 5D$ and (c) $r_{BA} = 10D$. In both cases the solid blue line represents the near field calculation and the dashed black line the far field calculation.

length scale of the swimming motion is typically $\sim 50D$, the trajectories evolve over timescales 3 or 4 orders of magnitude larger than that of the individual swimming stroke. The lines terminate when the swimmers get too close, as here the Oseen tensor approximation breaks down.

In addition to simple attractive or repulsive behaviour the swimmers also exhibit intriguing oscillatory trajectories which may either be of small amplitude about separated parallel swimming paths, as in region P, or of larger amplitude with the two swimmers' trajectories crossing each other, as in region O. These oscillations take place over the course of $\sim 10,000$ swimming cycles, although this period varies strongly with the separation of the two swimmers.

The type of trajectories observed depends on the relative phase of the swimmers. Fig. 6.4(b) shows the analogous diagram to Fig. 6.4(a), but with the two swimmers π out of phase. The trajectories change because the amplitude of the active rotation changes sign. The attractive region is replaced by a repulsive one. The oscillatory regime now produces oscillating parallel paths, an example of which is shown in Fig. 6.4(iv). Here the two swimmers follow parallel paths inclined at a small angle to the direction in which they would move in the absence of interactions. Both trajectories exhibit small amplitude, long period oscillations about the swimming direction; the oscillations of the two swimmers are exactly π out of phase, and the amplitude of the oscillations vanishes when the angle between them is ~30°, corresponding to one of the zeroes of Δn_{active} , Eq. (6.54).

Fig. 6.4(c) shows the intermediate case when the relative phase of the two swimmers is $\eta_{BA} = \pi/2$. This case is noteworthy as the amplitude for the lowest order part of the active rotation, Eq. (6.53), vanishes identically. Nonetheless, it is clear that the active terms still dominate over the passive interactions since the behaviour is different for swimmers placed in front or behind the central swimmer. This positional change is equivalent to changing the relative phase between the swimmers from $\pi/2$ to $3\pi/2$ and thus can only be important if the active terms are dominating the interaction.

The oscillations of Fig. 6.4(iii) are perfectly periodic and repeat indefinitely. This is a direct consequence of the self T-duality of the Najafi-Golestanian swimmer when the amplitudes of the two arms are equal. For swimmers that are not self T-dual, the oscillating states are no longer indefinitely stable. In particular, Fig. 6.4(v) shows the effect of making the front swimming amplitude, $\xi^{\rm F}$, smaller and the rear amplitude, $\xi^{\rm R}$, larger. Now the amplitude of the oscillations becomes successively larger until they can no longer be sustained and, finally, the two swimmers follow diverging trajectories. The opposite case, when $\xi^{\rm F} > \xi^{\rm R}$, is shown in Fig. 6.4(vi). The amplitude of the oscillations decays and the swimmers move closer together, with the lines terminating when the Oseen tensor approximation breaks down as a result of the swimmers getting too close.

In this Chapter we have described the interactions between two simple model linked-sphere swimmers. These interactions are not of the naively assumed dipolar form, nor do they originate from the average flow field generated by any given individual. Instead they arise from an active term, coming from the simultaneous efforts of both swimmers to swim. This results not only in a more rapid, r_{BA}^{-4} , decay with separation than would occur for dipolar interactions, but also in a sensitive dependence of the interactions to the relative phase between the two swimmers, a feature recognised as important by Taylor [114], but not explicitly incorporated into many minimal swimmer models. In the next Chapter we shall investigate the form of the interactions between two swimmers further by considering swimmer scattering, and will find again that the relative phase plays an important role.



Figure 6.4: Long time behaviour of two Najafi-Golestanian swimmers, (a) in phase, (b) π out of phase, (c) $\eta_{BA} = \pi/2$, and (d) in phase, but with $\xi^{\rm F} > \xi^{\rm R}$. (i)–(vi) illustrate a selection of trajectories representing attractive (A), repulsive (R), oscillatory (O), parallel (P), expanding oscillatory (OE), and contracting oscillatory (OC) behaviour, respectively.

CHAPTER 7

Swimmer scattering

Consider two swimmers that are approaching each other in the fluid. As they approach the hydrodynamic interactions between them will cause them both to rotate and subsequently to separate along new trajectories. This classic scenario describes a scattering event and provides the focus of our attention for this Chapter.

7.1 Geometry of scattering

In a generic scattering event the two swimmers, A and B, have instantaneous positions $\mathbf{x}^{A}(t), \mathbf{x}^{B}(t)$ and swimming directions $\mathbf{n}^{A}(t), \mathbf{n}^{B}(t)$. We use these positions and orientations to describe the configuration of the swimmers via a simple geometric construction, shown in Fig. 7.1. The position and orientation of each swimmer generates a straight line. Provided the two swimming directions are not parallel, there will be unique points, \mathbf{l}^{A} and \mathbf{l}^{B} , at which they are closest to each other. We denote by s^{A}, s^{B} the distances of each swimmer from these points of closest approach, taken positive if \mathbf{l} is ahead of the swimmer and negative otherwise. The difference between these two distances, $b := s^{A} - s^{B}$, is called the *impact parameter* and measures how closely the two swimmers would approach each other in the absence of any interactions. The minimum distance between the two lines, $l := |\mathbf{l}^{A} - \mathbf{l}^{B}|$, is called the *lift parameter* and the angle between the two swimmers is called the *angle of incidence*, $\alpha := \arccos(\mathbf{n}^{A} \cdot \mathbf{n}^{B})$. The limiting values of these quantities are used to define the initial and final states

$$\left(\alpha_{\pm}, b_{\pm}, l_{\pm}\right) = \lim_{t \to \pm \infty} \left(\alpha(t), b(t), l(t)\right) \,. \tag{7.1}$$


Figure 7.1: Schematic diagram of the geometry of a scattering event: see text for details. For ease of illustration we show a planar event, i.e., l = 0.

Finally, the change in orientation of an individual swimmer as a result of the scattering process is described by the *scattering angle*

$$\theta = \arccos\left(\mathbf{n}(t \to -\infty) \cdot \mathbf{n}(t \to +\infty)\right) \,. \tag{7.2}$$

7.2 Scattering trajectories

The scattering trajectories of two Najafi-Golestanian swimmers can be determined using the results presented in Chapter 6. We briefly describe the general features, for planar scattering geometries, l = 0, which set the scene for this Chapter. Three generic types of scattering event are observed: the swimmers can *collide*, by which we mean they approach so closely that the Oseen tensor approximation is no longer appropriate, they can *exchange trajectories*, or they can *turn in the same direction*. Exemplary trajectories for the latter two types of events are shown in Fig. 7.2. In the turn event the impact parameter is relatively large and swimmer B is able to pass directly in front of swimmer A, causing both of them to rotate in the clockwise sense. For the exchange event, the impact parameter is slightly smaller, and in this case the swimmers do not cross, but rotate in opposite directions, essentially swapping places. The remarkable feature of both events is that the angle between the swimmers is unchanged by the scattering.



Figure 7.2: Trajectories of two Najafi-Golestanian swimmers during planar scattering. (I) a turn event, and (II) an exchange event.

7.3 T-duality and swimmer scattering

In this Section we shall describe the consequences of the time reversal invariance of the Stokes equations for the hydrodynamic scattering of two low Reynolds number swimmers, and argue that this provides the explanation for the preservation of angle described in § 7.2.

Hydrodynamic scattering may be viewed as providing a map from the initial state (α_-, b_-, l_-) to the final state (α_+, b_+, l_+) . The differences between these two states, defined by the functions $\delta_{\alpha} := \alpha_+ - \alpha_-$, $\delta_b := b_+ - b_-$ and $\delta_l := l_+ - l_-$, describe the tendancy for the swimmers to align $(\delta_{\alpha} < 0)$ or cluster $(\delta_b < 0, \delta_l < 0)$ via hydrodynamic interactions. Viewing the entire process backwards in time corresponds to the hydrodynamic scattering of the T-dual swimmers (\bar{B}, \bar{A}) taking the initial state (α_+, b_+, l_+) into the final state (α_-, b_-, l_-) , thereby establishing an isomorphism between the scattering of an arbitrary pair of swimmers and the scattering of their T-duals. In particular the functions $\delta^{(\bar{B},\bar{A})}$ are simply related to the functions $\delta^{(A,B)}$

$$\delta^{(\bar{B},\bar{A})}(\alpha_{+},b_{+},l_{+}) = -\delta^{(A,B)}(\alpha_{-},b_{-},l_{-}).$$
(7.3)

In the case of a pair of mutually T-dual swimmers $(\bar{B} = A; \bar{A} = B)$ this is sufficient to show that $(\alpha_+, b_+, l_+) \equiv (\alpha_-, b_-, l_-)$.

We motivate this result using symmetry arguments. During any scattering event the quantity $s^A + s^B$ changes from being large and positive to being large and negative. Since it does this continuously it must pass through zero, which we use to define the time t = 0. The separation



Figure 7.3: Time reversal transformations and swimmer scattering. At t = 0 reversing the direction of time and performing a π rotation about the axis indicated returns an identical configuration if the pair are mutually T-dual. Top row: a turn event. Bottom row: an exchange event.

between the two swimmers is given by

$$\mathbf{r}_{BA} = \frac{1}{2} \left(s^A + s^B \right) \left(\mathbf{n}^A - \mathbf{n}^B \right) + \frac{1}{2} \left(s^A - s^B \right) \left(\mathbf{n}^A + \mathbf{n}^B \right) + \left(\mathbf{l}^B - \mathbf{l}^A \right) , \qquad (7.4)$$

and is orthogonal to the direction $\mathbf{n}^A - \mathbf{n}^B$ at t = 0. At this instant reversing the direction of time, followed by a π rotation about an axis parallel to $\mathbf{n}^A - \mathbf{n}^B$ and passing through the point mid-way between the two swimmers leads to a configuration where $\overline{B}, \overline{A}$ have the same positions and orientations as A, B, respectively. As illustrated in Fig. 7.3, for mutually T-dual swimmers this returns the same configuration we started with. It follows that A's outgoing trajectory for t > 0 will be given by B's ingoing trajectory for t < 0 (with the direction of time reversed) and vice-versa, from which we conclude that the initial and final states are the same. In addition, for planar scattering geometries, this construction implies that the swimmers rotate in the same direction and with equal scattering angles, $\theta^A = \theta^B$. These are the *turn* events, of which an example was shown in Fig. 7.2(I).

An exception to this scenario occurs if the swimmers ever become exactly parallel, $\mathbf{n}^A = \mathbf{n}^B$. However, taking t = 0 at this instant and choosing the rotation axis to be parallel to $\mathbf{n}^A \times \mathbf{r}_{BA}$ leads to the same conclusion (Fig. 7.3). This time, since the two swimmers rotate in opposite directions, the constraint that $\alpha_+ = \alpha_-$ can only be met if the scattering angles take the values $\theta = \pm \alpha_-$ independent of b_- . In such an event A will rotate so that its outgoing trajectory is parallel to B's ingoing trajectory and vice-versa. These are the exchange events, an example of which was shown in Fig. 7.2(II). Since we expect $\theta \to 0$ as $b_- \to \infty$, exchange events can only occur for sufficiently small values of b_- . Finally, we comment that, since for purely planar scattering there is no way to cross smoothly between these two cases, they are necessarily separated by some form of discontinuous behaviour. We speculate that the swimmers enter a trapped, or bound, state from which they are unable to escape.

There are a number of subtleties in the foregoing observations. To exactly interchange the swimmers as described, the time t = 0 must coincide with particular instants during the swimming cycle, otherwise, although the positions and orientations of the swimmers will be the same after the interchange, the stages they are at during their swimming strokes will not. We have not been able to show generally that the time t = 0 does indeed coincide with one of these instances, although for this not to be the case would imply rather peculiar properties for the functions $\delta^{(A,\bar{A})}$. Also, since our discussion has mentioned only the swimming direction, **n**, it has been restricted to swimmers that are axisymmetric, requiring only this vector to completely specify their orientation.

Moreover, we have defined the time t = 0 through the condition that the vector $\mathbf{n}^A - \mathbf{n}^B$ is orthogonal to the relative position vector \mathbf{r}_{BA} of the two swimmers, but we have not defined what this means. In particular, what is the relative position of the two swimmers? Is it the separation between the two centre spheres? Or between the two leading spheres (the 'head' of the swimmer)? In fact, it does not matter which point (i.e., which sphere) is chosen to represent the position of a given swimmer, a consequence of the insignificance of inertia at low Reynolds number [119]. Different choices correspond to a change of 'gauge' and invariance of the single swimmer motion under gauge transformations leads to the beautiful gauge field theory of swimming introduced by Shapere and Wilczek [118–120].

But different choices do matter here. If the vector $\mathbf{n}^A - \mathbf{n}^B$ is orthogonal to the relative position vector of the two centre spheres it will not simultaneously be orthogonal to the relative position vector of the two 'head' spheres. Nonetheless, a unique definition of t = 0 is still possible. $\mathbf{n}^A - \mathbf{n}^B$ cannot be simultaneously orthogonal to the relative positive vectors of all pairs of points on the two swimmers, but it can be simultaneously orthongal to all pairs of *conjugate* points. Conjugate points are defined between a swimmer and its T-dual such that they correspond to the same material point on the two swimmers. This is most easily illustrated by a figure, as we have done in Fig. 7.4. The condition that the relative position vectors of



Figure 7.4: The relationship between a swimmer and its T-dual naturally introduces the notion of conjugate points. The foremost part of a swimmer is conjugate to the rearmost part of its T-dual and vice-versa.

all pairs of conjugate points are simultaneously orthogonal to the vector $\mathbf{n}^A - \mathbf{n}^B$ is sufficient to guarantee that, after the interchange described above, \bar{B} has the same position, orientation and is also at the same stage through the swimming stroke, as A before the interchange.

In numerical tests using both the Najafi-Golestanian model and a model of snake-like swimmers, the swimmers do indeed reach t = 0 at a suitable stage of their stroke. This is shown in Fig. 7.5 for both turn and exchange type events with the Najafi-Golestanian swimmer. Under T-duality, the rear arm of swimmer A becomes the front arm of swimmer $\bar{A} = B$. Thus, for the interchange of the two swimmers to be exact it must occur at a stage through the swimming stroke where the rear arm of A has the same extension as the front arm of B. Parameterising the swimming stroke by a variable $\tau \in [0, 2\pi)$, we see that this implies the condition

$$\sin(\tau) = \sin(\tau + \eta_{BA} - \phi), \qquad (7.5)$$

which is satisfied for $\tau = ((2m+1)\pi + \phi - \eta_{BA})/2$, with m = 0, 1. In the simulations described here, the swimmers were in phase $\eta_{BA} = 0$ and the phase lag between front and rear arm oscillations was $\phi = \pi/2$. For these values the interchange is exact only if $\tau = 3\pi/4$ or $\tau = 7\pi/4$. As can be seen in Fig. 7.5, for the turn event the vector $\mathbf{n}^A - \mathbf{n}^B$ is orthogonal to \mathbf{r}_{BA} for all three pairs of conjugate points simultaneously at a stage corresponding exactly to $\tau = 7\pi/4$, while for the exchange event the two swimmers are parallel at a stage corresponding exactly to $\tau = 3\pi/4$.



Figure 7.5: Numerical identification of the time t = 0 in the scattering of a pair of mutually T-dual swimmers, verifying the exact interchange. The black arrows indicate the start of swimming stroke of swimmer A, each swimming stroke corresponding to 80 simulation timesteps. For the turn event the three sets of data correspond to the three choices of conjugate points to measure the relative position \mathbf{r}_{BA} . These are: the tail sphere of A and head sphere of B (red), the two centre spheres (blue), and the head sphere of A and the tail sphere of B (cyan). I am grateful to Vic Putz for providing these figures.

7.4 Scattering of the Najafi-Golestanian swimmer

These general symmetry considerations provide a framework for what can be expected in two body swimmer scattering. In the remainder of this Chapter we illustrate and extend the results by describing in more detail the planar hydrodynamic scattering of two Najafi-Golestanian swimmers. We consider first two identical swimmers that are in phase, $\eta_{BA} = 0$, and have equal arm amplitudes, $\xi^{R} = \xi^{F}$, for which the swimming stroke is self T-dual. For all trajectories $(\alpha_+, b_+) = (\alpha_-, b_-)$ in accordance with the symmetry arguments we have presented above. The type of scattering event (exchange or turn) that occurs is shown as a function of the two initial conditions α_{-}, b_{-} in Fig. 7.6(a), together with a detailed cut showing how the scattering angle θ varies with b_{-} for a fixed value of $\alpha_{-} = 30^{\circ}$ (Fig. 7.6(b)). There is a wide range of initial conditions at small values of b_{-} for which the scattering is of the exchange type and $\theta = \pm \alpha_{-}$. At larger values of b_{-} the scattering is always of the turn type with the scattering angle decaying to zero as $b_{-} \rightarrow \infty$. In the region labelled Collide the swimmers approach so closely that the Oseen tensor description of the hydrodynamics is no longer valid and we are unable to determine what happens during the scattering. In our simulations we took this to occur when the minimum separation between any two spheres became less than 10a = 0.5D.

The behaviour is similar for different values of the relative phase between the swimmers. In



Figure 7.6: Hydrodynamic scattering of two identical, self T-dual Najafi-Golestanian swimmers. (a) and (c) The type of scattering observed for different values of the initial conitions α_{-}, b_{-} when the swimmers are in phase, $\eta_{BA} = 0$, and exactly out of phase, $\eta_{BA} = \pi$, respectively. (b) and (d) Dependence of the scattering angle on b_{-} for a fixed value of $\alpha_{-} = 30^{\circ}$, corresponding to the dashed lines in (a) and (c), respectively.

Fig. 7.6(c) and (d) we show the results for two swimmers that are exactly out of phase, $\eta_{BA} = \pi$. The same types of scattering are found, but with the boundaries between the different regions shifted to smaller values of b_- . An interesting feature is that the scattering angle in the turn regime does not decay to zero monotonically with increasing b_- as is the case for swimmers that are in phase. At large values of b_- the scattering angle is negative for $\eta_{BA} = 0$ and positive for $\eta_{BA} = \pi$, reflecting the change in sign of the amplitude of the far field rotational interaction as given by Eq. (6.53). If the far field approximation to the hydrodynamics provided a sufficient description of the interactions we might expect the scattering angle to be positive for all $b_$ when the swimmers are exactly out of phase, i.e., simply the opposite sign to when they are in phase. That this is not the case reflects the need to capture the interactions more accurately when the separation between the swimmers is comparable to their size.

We now outline how the properties of swimmer scattering change when the two swimmers are not mutually T-dual and our preceeding symmetry arguments no longer apply. In Fig. 7.7(a) we show the change in alignment δ_{α} as a function of b_{-} for both a pair of identical extensile swimmers, with $(\xi^{\rm R}, \xi^{\rm F}) = (0.3D, 0.1D)$, and a pair of identical contractile swimmers, with $(\xi^{\mathrm{R}},\xi^{\mathrm{F}}) = (0.1D, 0.3D)$, with a relative phase of $\eta_{BA} = \pi/2$ in both cases. Not only is δ_{α} nonzero, it is generically large; several tens of degrees in many cases. For this choice of relative phase we see a predominant tendancy for extensile swimmers to increase the angle between the two swimmers, i.e., $\delta_{\alpha} > 0$, while contractile swimmers predominantly show hydrodynamically induced alignment, $\delta_{\alpha} < 0$.

However, the relative phase is found to play an important role here. We show in Fig. 7.7(b) the change in alignment δ_{α} as a function of the relative phase for fixed $\alpha_{-} = 30^{\circ}$, $b_{-} = 4D$ and the same two pairs of swimmers. Several features are noteworthy. The most striking feature is the appearance of an extended region where $\delta_{\alpha} = -30^{\circ}$ for the pair of contractile swimmers. This represents the formation of a bound state in which the two swimmers are exactly aligned one behind the other. An example of swimmer trajectories during the formation of this bound state is shown in Fig. 7.7(d). A smaller region exhibiting the same behaviour can also be seen in Fig. 7.7(a).

As the relative phase is varied, δ_{α} is as often positive as it is negative, for both extensile and contractile pairs of swimmers. Thus there is not a simple, direct relationship between the change in angle between the two swimmers and their being contractile or extensile. However, the behaviour is asymmetric in the sense that the magnitude of δ_{α} is larger for $0 < \eta_{BA} < \pi$ than for $\pi < \eta_{BA} < 2\pi$ in both cases. Moreover, there are two values of the relative phase, $\eta_{BA} = 0$ and $\eta_{BA} = \pi$, for which $\delta_{\alpha} = 0$. Thus we find an intriguing result: when the swimmers are exactly in phase, or exactly out of phase, hydrodynamic scattering does not change the angle between them, even if they are not mutually T-dual. It is tempting to speculate that this might also follow from symmetry considerations, or that it might apply to swimmers other than the Najafi-Golestanian model considered here.

On either side of the bound state regions in both Fig. 7.7(a) and (b), δ_{α} takes a value somewhat larger than -30° , indicating that δ_{α} is discontinous at the transition from scattering to bound state formation. This qualitative observation is supported by the behaviour when the transition is approached from a different direction, that of increasing the difference in amplitudes $\xi^{\rm F} - \xi^{\rm R}$ at a fixed value of $b_- = 4D$ as shown in Fig. 7.7(c). We vary this difference in amplitudes whilst holding the product $\xi^{\rm R}\xi^{\rm F}$ fixed to maintain an approximately constant swimming speed, Eq. (6.13), [140, 143, 166]. As $\xi^{\rm F} - \xi^{\rm R}$ is increased from zero δ_{α} decreases smoothly until it reaches the value 0.16D where there is an abrupt transition to the bound state, accompanied by a substantial discontinuity in δ_{α} .



Figure 7.7: Hydrodynamic scattering of Najafi-Golestanian swimmers that are not mutually T-dual. The change in angle between two swimmers as a function of (a) the impact parameter for a fixed relative of phase $\eta_{BA} = \pi/2$, and (b) their relative phase at a fixed value of the impact parameter, $b_{-} = 4D$. In both cases the red circles correspond to a pair of identical extensile swimmers with $\xi^{\rm R} = 0.3D, \xi^{\rm F} = 0.1D$ and the blue crosses correspond to a pair of identical contractile swimmers with $\xi^{\rm R} = 0.1D, \xi^{\rm F} = 0.3D$. In (c) we show how the change in angle between the swimmers depends on the amplitude asymmetry $\xi^{\rm F} - \xi^{\rm R}$ for $b_{-} = 4D$ and $\eta_{BA} = \pi/2$. In (d) we show an exemplary scattering trajectory leading to the formation of a bound state for $\xi^{\rm F} - \xi^{\rm R} = 0.17D$ and other parameters as in (c).

7.5 Discussion

We have described the constraints imposed on the hydrodynamic scattering of two swimmers by the time reversal invariance of the Stokes equations. The most striking observation concerns Tdual swimmers, which have strokes that map onto each other under time reversal: for scattering events involving two such swimmers the angle and impact parameter between their trajectories are the same before and after the collision. For swimmers unrelated by T-duality we show numerically that scattering is complex, with changes in the angle between the two swimmers of several tens of degrees, and allowing the possibility of forming bound states. Experiments on biological or fabricated microswimmers should show these striking differences between pairs of mutually T-dual and symmetry unrelated swimmers.

In future work it would be particularly interesting to extend our ideas to more than two swimmers. In suspensions of self T-dual swimmers the evolution of the system may be constrained to look the same both forwards and backwards in time, at least in a statistical sense, and it may be possible to exploit this to obtain exact results for their non-equilibrium dynamics. Furthermore, these microscopic symmetries of the interactions between individuals should also appear in the coarse-grained hydrodynamic equations of continuum theories. It will be important to determine in what way self T-duality enters into the continuum equations and how it effects the resulting macroscopic behaviour.

The description of the scattering of two swimmers described in this Chapter is not yet a complete one and there are many aspects that merit further study. Determining the limiting behaviour of the swimmers during the discontinuous cross-over from turn to exchange type events is one such unresolved problem, but this is beyond the validity of the Oseen tensor approach. Another is the status of non-axisymmetric swimmers; it is not yet clear whether or not they exhibit the same exact recovery of the initial state in the long time limit as we described for axisymmetric swimmers. It is tempting to speculate that they do, but this still has to be shown, either generally or at least for some specific cases. And this brings us to our final comment. A proof of the remarkable scattering behaviour described in this Chapter is still needed; we have only been able to provide part of it. It is both interesting and important to complete it.

CHAPTER 8

Dumb-bell swimmers

In the low Reynolds number world of microorganisms, reciprocal strokes do not lead to swimming [116, 117, 169]. Organisms that perform such reciprocal motions, referred to as *apolar* swimmers or "shakers" [109], are able to generate fluid flow but unable to generate their own motility. However, as pointed out by Koiller *et al* [172], although this is true for a single apolar swimmer, a collection of such swimmers *can* swim. If their swimming strokes are not in phase, the motion taken as a whole is not reciprocal in time, and motility is possible. The physical mechanism which turns the motion of an apolar swimmer into a net displacement is the hydrodynamic forces between the swimmers.

In this Chapter we study the simplest model of an apolar swimmer, oscillating dumb-bells. Following the approach we developed in Chapter 6, we give an analytic solution for the motion of a pair of dumb-bells, in the Oseen tensor limit and valid for large separations. By numerically iterating these results we are able to gain insight into how the orbits of two swimmers depend on their relative positions, separations, and phases.

The motility of an apolar swimmer is inherently collective: a single dumb-bell does not swim, but more than one can. It is natural, therefore, to investigate how this collective locomotion develops as the number of dumb-bells is increased. We do this by first considering simple systems, principally linear chains and regular arrays, and describe how the resulting motion can be understood in terms of the pairwise interactions between individuals.

Finally we describe the motion of a suspension of randomly placed and randomly oriented dumb-bells. Since each dumb-bell is intrinsically apolar, a suspension of them provides a minimal microscopic model of an active apolar fluid. These active fluids have received a lot of interest in the past decade [98,99,108,109,173–175], both as examples of driven non-equilibrium

systems and because of their close analogy to active polar fluids [98,99,176–178]. The properties of such systems have been formulated primarily at the continuum level [98,99], on the basis of symmetry and conservation laws, but have also been studied numerically for suspensions of individuals interacting through simple modelling rules [174].

8.1 Hydrodynamic interactions between oscillating dumb-bells

We first describe the motility of oscillating dumb-bells at zero Reynolds number. Each dumbbell comprises two spheres, of radius a, joined by a thin, rigid rod, whose length varies sinusoidally as $D + \xi \sin(\omega t)$. As in other linked sphere models the rod is treated as a phantom linker and its hydrodynamic effect neglected. We stress the importance of the relative phase of the two dumb-bells: if they oscillate in phase or π out of phase then their combined movements remain reciprocal and the Scallop theorem prevents any net motion [117]. However, for other values of the relative phase, motion will occur. To show this we use the Oseen tensor formulation of hydrodynamics, valid in the limit of zero Reynolds number [170], with an analysis similar to Chapter 6. We present anaytic calculations, valid for large separations, and numerical results based on a more detailed near field calculation like that described in § 6.3.4, valid for separations down to of order the size of a dumb-bell.

As described in Chapter 6, linearity of the Stokes equations allows the fluid velocity to be written as a linear combination of the forces acting on the fluid due to the motion of the dumb-bells

$$\mathbf{u}(\mathbf{x}) = \sum_{A} \left[\mathbf{G}_{A}^{1}(\mathbf{x}) \mathbf{f}_{A}^{1} + \mathbf{G}_{A}^{2}(\mathbf{x}) \mathbf{f}_{A}^{2} \right].$$
(8.1)

Here the subscript A labels the dumb-bells, the superscripts 1,2 label the spheres of an individual dumb-bell and the Green function is again taken to be the Oseen tensor, Eq. (6.5), assuming $a/D \ll 1$. The motion of the dumb-bells through the fluid is determined, as before, by three ingredients; consistency of the fluid flow with the change in shape, i.e., the change in rod length, the constraint that each dumb-bell is force-free, $\mathbf{f}_A^1 + \mathbf{f}_A^2 = \mathbf{0}$, and the constraint that each dumb-bell is torque-free, $[(\mathbf{x}_A^2 - \mathbf{x}_A^1), \mathbf{f}_A^1] = \mathbf{0}$. A short calculation then leads to an expression for the forces

$$\mathbf{f}_{A}^{1} = -3\pi\mu a \left\{ \xi\omega \cos(\omega t) \left(1 + \frac{3}{2} \frac{a}{D + \xi \sin(\omega t)} \right) \mathbf{n}_{A} + \left(D + \xi \sin(\omega t) + \frac{3a}{4} \right) \left[\Omega_{A} \,, \, \mathbf{n}_{A} \right] + \dots \right\} \\ + 3\pi\mu a \sum_{B \neq A} \left\{ \mathbf{G}_{B}^{1}(\mathbf{x}_{A}^{2}) - \mathbf{G}_{B}^{1}(\mathbf{x}_{A}^{1}) - \mathbf{G}_{B}^{2}(\mathbf{x}_{A}^{2}) + \mathbf{G}_{B}^{2}(\mathbf{x}_{A}^{1}) \right\} \mathbf{f}_{B}^{1} , \qquad (8.2)$$

where terms of $O[(a/D)^2]$ have been omitted. The unit vector \mathbf{n}_A gives the direction of sphere 2 relative to sphere 1 and thus describes the orientation of the dumb-bell. As in Chapter 1 we describe an apolar quantity by a vector instead of a line element purely for mathematical simplicity, but, of course, our results must be independent of this artificial orientation assigned to a dumb-bell. Applying the torque-free constraint determines the angular velocity, Ω_A , from which we obtain an equation for the evolution of the dumb-bell's orientation

$$\frac{\mathrm{d}\mathbf{n}_A}{\mathrm{d}t} = \left[\Omega_A \,,\, \mathbf{n}_A\right] \,,\tag{8.3}$$

$$= \frac{1}{D+\xi\sin(\omega t)} \Big[\mathbf{I} - \mathbf{n}_A \otimes \mathbf{n}_A \Big] \sum_{B \neq A} \Big\{ \mathbf{G}_B^1(\mathbf{x}_A^2) - \mathbf{G}_B^1(\mathbf{x}_A^1) - \mathbf{G}_B^2(\mathbf{x}_A^2) + \mathbf{G}_B^2(\mathbf{x}_A^1) \Big\} \mathbf{f}_B^1 .$$
(8.4)

Finally, the translational motion of each dumb-bell is given by

$$\frac{\mathrm{d}\mathbf{x}_{A}^{c}}{\mathrm{d}t} = \frac{1}{2} \sum_{B \neq A} \left\{ \mathbf{G}_{B}^{1}(\mathbf{x}_{A}^{2}) + \mathbf{G}_{B}^{1}(\mathbf{x}_{A}^{1}) - \mathbf{G}_{B}^{2}(\mathbf{x}_{A}^{2}) - \mathbf{G}_{B}^{2}(\mathbf{x}_{A}^{1}) \right\} \mathbf{f}_{B}^{1} , \qquad (8.5)$$

where \mathbf{x}_A^c denotes the 'centre' of the dumb-bell, $\mathbf{x}_A^c := (\mathbf{x}_A^1 + \mathbf{x}_A^2)/2$. As expected, the motion described by Eqs. (8.4) and (8.5) arises solely through interactions with other dumb-bells.

We assume that the dumb-bells are in a dilute suspension so that the separation, r, of any given dumb-bell from its nearest neighbour may be assumed to be large compared to its size, D. Under such circumstances the interactions between the dumb-bells may be expanded in a power series in (D/r) and only the leading contributions retained. Integrating over a complete cycle leads to expressions for the changes in position and orientation of the dumb-bells after a single swimming stroke:

$$\Delta \mathbf{n}_{A} = \sum_{B \neq A} \frac{15a\mathcal{A}}{32 r_{BA}^{5}} \Big(\hat{\mathbf{r}}_{BA} - (\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA}) \mathbf{n}_{A} \Big) \Big\{ 3(\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA}) + 6(\mathbf{n}_{A} \cdot \mathbf{n}_{B})(\mathbf{n}_{B} \cdot \hat{\mathbf{r}}_{BA}) + 6(\mathbf{n}_{A} \cdot \mathbf{n}_{B})^{2} (\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA}) - 7(\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA})^{3} - 21(\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA})(\mathbf{n}_{B} \cdot \hat{\mathbf{r}}_{BA})^{2} - 42(\mathbf{n}_{A} \cdot \mathbf{n}_{B})(\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA})^{2} (\mathbf{n}_{B} \cdot \hat{\mathbf{r}}_{BA}) + 63(\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA})^{3} (\mathbf{n}_{B} \cdot \hat{\mathbf{r}}_{BA})^{2} \Big\},$$

$$(8.6)$$



Figure 8.1: Orbits of a pair of coplanar dumb-bells. The two dumb-bells are initially parallel with A at the origin and B placed at the corresponding point in the diagram. Several regimes of long time behaviour are found. In region P the two dumb-bells adopt a stable perpendicular configuration as exemplified by (i). This type of behaviour is also found in the shaded regions, however, before acquiring the perpendicular configuration the two dumb-bells tumble for a period of time, as shown in (ii). In the regions labelled O the dumb-bells move along parallel trajectories with an oscillatory motion, as illustrated in (iii). Finally, along each of the dashed lines there is no rotational interaction and the dumb-bells undergo a pure translation along straight parallel paths.

$$\Delta \mathbf{x}_{A}^{c} = \sum_{B \neq A} \frac{9a\mathcal{A}}{32 r_{BA}^{4}} \left\{ \hat{\mathbf{r}}_{BA} \left[1 + 2(\mathbf{n}_{A} \cdot \mathbf{n}_{B})^{2} - 5(\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA})^{2} - 5(\mathbf{n}_{B} \cdot \hat{\mathbf{r}}_{BA})^{2} - 20(\mathbf{n}_{A} \cdot \mathbf{n}_{B})(\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA})(\mathbf{n}_{B} \cdot \hat{\mathbf{r}}_{BA}) + 35(\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA})^{2}(\mathbf{n}_{B} \cdot \hat{\mathbf{r}}_{BA})^{2} \right] + 2\mathbf{n}_{A} \left[(\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA}) + 2(\mathbf{n}_{A} \cdot \mathbf{n}_{B})(\mathbf{n}_{B} \cdot \hat{\mathbf{r}}_{BA}) - 5(\mathbf{n}_{A} \cdot \hat{\mathbf{r}}_{BA})(\mathbf{n}_{B} \cdot \hat{\mathbf{r}}_{BA})^{2} \right] \right\},$$

$$(8.7)$$

where \mathbf{r}_{BA} is the position vector of B relative to A. The amplitude A is given by

$$\mathcal{A} = \pi D^2 \xi_A \xi_B \sin(\eta_{BA}) + \frac{\pi}{8} \xi_A^2 \xi_B^2 \sin(2\eta_{BA}) , \qquad (8.8)$$

where η_{BA} is the phase of *B*'s swimming stroke relative to *A*'s. The long time behaviour of a group of dumb-bells may be determined by numerically iterating Eqs. (8.6) and (8.7) to find the new positions and orientations of all the dumb-bells after each swimming cycle.

8.2 Two dumb-bells

We first consider two dumb-bells lying in the yz-plane and both oriented along the z-direction. Dumb-bell A is initially at the origin and dumb-bell B is placed on a circle of radius 4D centred on the origin. For two dumb-bells, varying the relative phase does not lead to any qualitative changes in behaviour and therefore we consider only the case $\eta_{BA} = \pi/2$. The sole free parameter is the angle, θ , that the position vector of B makes with the y-direction. As this angle is varied the hydrodynamic interactions between the dumb-bells change leading to different long time behaviour, which we illustrate in Fig. 8.1.

The predominant behaviour is for the two dumb-bells to adopt a perpendicular configuration in which one dumb-bell is oriented parallel and the other perpendicular to their relative position vector. This stable arrangement appears at long times for all initial configurations in the regions labelled P in Fig. 8.1. An exemplary time series showing how the perpendicular configuration is reached is shown in Fig. 8.1(i). The stability of this state may be seen from a linear stability analysis of Eq. (8.6).

Similarly, linear stability analysis reveals that the rotational fixed point at $\theta = 0$ is unstable. However, small deviations away from this fixed point do not lead smoothly to the stable perpendicular configuration. Instead there is an initial period during which the dumb-bells tumble, often several times, before they finally settle down, as illustrated in Fig. 8.1(ii). Exactly at the fixed point, and in the absence of any fluctuations, the dumb-bells undergo a pure translational motion, swimming cooperatively in the direction of the dumb-bell with positive relative phase.

The fully aligned configuration with $\theta = \pi/2$ is also a rotational fixed point and again, exactly at this angle, the motion is purely translational with both dumb-bells moving in the same direction. However, in this case, the fixed point is a centre and small deviations away from it lead to an oscillatory cooperative motion, an exemplary time series of which is shown in Fig. 8.1(iii). This oscillatory motion occurs throughout the regions labelled O in Fig. 8.1 and is separated from the P regions by an additional rotational fixed point at $\theta \approx 65^{\circ}$.

8.3 Linear chains of dumb-bells

We now present examples of the cooperative effects of the hydrodynamic interaction between many apolar swimmers, for first regular, and then random, distributions of swimmers. Consider a chain of N identical dumb-bells all oriented along the z-direction and initially positioned at equally spaced intervals of 5D along the y-axis. To optimise the hydrodynamic interactions between nearest neighbours the relative phase between any two neighbouring dumb-bells is set



Figure 8.2: (a) Configuration of a linear chain of dumb-bells and a schematic representation of its evolution. The bulk of the chain moves faster than the boundary, leading to dumb-bells being left behind at the trailing boundary and the formation of 'fast pairs' at the leading boundary. (b) The distances moved by each dumb-bell in the chain after 100,000 swimming strokes for chains with different numbers of dumb-bells.

to $\pi/2$, increasing in the positive y-direction. Although this is a highly artificial configuration, the lack of any rotation greatly simplifies the dynamics, allowing for the effect of changing N to be clearly quantified.

We show in Fig. 8.2 the initial configuration of the chain and the distances moved by each of the dumb-bells as their total number increases. Two features are particularly noteworthy: firstly the behaviour for N > 2 is significantly different from that for N = 2, and secondly the general behaviour for N > 20 shows only minor variations, indicating that an asymptotic limit is being approached.

The evolution of the pattern of swimmers may be understood by noting that there is a fundamental distinction between dumb-bells with two nearest neighbours and those with only one. The former constitute what we shall call the *bulk* of the chain, while the latter form the *boundary*. A dumb-bell in the bulk not only gets pulled along by its neighbour in front of it, but is also pushed by its neighbour from behind. These two interactions add constructively leading to a member of the bulk moving faster than a single isolated pair. By contrast a dumb-bell which is on the boundary only has one nearest neighbour and hence does not benefit from this added boost. Thus the bulk moves faster than the boundary.

This has the effect of introducing an asymmetry between the two boundaries; the dumbbell on the trailing boundary gets left behind while that on the leading boundary is caught up. From Eq. (8.7) we see that the strength of interactions depends on the separation of the dumb-bells as r^{-4} , so that as the trailing dumb-bell gets left behind, the pull it receives from the bulk rapidly diminishes until it becomes isolated and can no longer move. At the same time the distance between the leading dumb-bell and the bulk decreases yielding a sharp increase in the strength of interaction between itself and its neighbour. As a result the leading pair speed up significantly and are ejected from the front of the chain.

8.4 Lattice pumps

The cooperative motion of one dimensional chains carries over to regular arrays in two dimensions. As an example, consider a square lattice of dumb-bells all oriented in the z-direction and with (x, y) positions (jL, kL), where L is the lattice constant and j, k are integers. Co-operative directed motion can be induced by defining the phase of each dumb-bell to be $\phi_{j,k} = \pi (j+k)/2$. The entire lattice then moves uniformly along the [110] direction.

For a system of real swimmers this state could not be sustained; we have found that it is unstable to any imperfections of the lattice such as boundaries or fluctuations in the position of the swimmers. This instability of long range coherent states is in agreement with analytic work by Ramaswamy and co-workers [98,99,108] and numerical simulations of Saintillan and Shelly [113]. However, one might envisage tethering fabricated dipolar swimmers to a substrate and aligning and activating them with a magnetic field. Such a set-up would act as a miconscale pump. We estimate $v \sim \mu m s^{-1}$, which is similar to the velocities achieved by bacteria. It should be noted that the interactions scale with separation as r^{-4} , so that a small decrease in lattice spacing will provide a substantial increase in flow speed.

8.5 Suspensions of dumb-bells

We next consider a suspension of N dumb-bells initially dispersed throughout a cubic box of side L = 20D with random positions, orientations and relative phases. To avoid singularities in the hydrodynamic interactions a short distance cut-off is employed when the separation between any two dumb-bells becomes less than 0.5D. At separations of order D an expansion of the Oseen tensor in powers of D/r does not converge rapidly. To overcome these difficulties we instead use an expansion in powers of ξ/r to better describe the near field hydrodynamics.

The combination of a random initial configuration and apolar symmetry means that there is no prefered direction for the motion, and on average the velocity is zero. In continuum models of apolar active fluids spontaneous symmetry breaking can lead to a state with non-zero average velocity [175, 176], however we have not observed any such transitions in our simulations of



Figure 8.3: Collective properties of a suspension of interacting (I) dumb-bell swimmers (apolar) and (II) three-sphere swimmers (polar). Left: mean speed of the swimmers as a function of their number density. The line indicates a linear fit. Centre: mean square displacement as a function of time for number densities $0.1 (\times)$ and $0.02 (\diamond)$. Right: diffusion constant for increasing number of swimmers.

dumb-bells. In the absence of a net velocity, the mean speed provides a measure of the degree of collective activity. Fig. 8.3(I) shows that the mean speed of the dumb-bells increases linearly with the number density $n = N/L^3$. This agrees with a simple scaling argument: since every dumb-bell will interact with every other one, the total number of interactions scales as n^2 . Balancing this against the rate of dissipation of energy predicts that the mean speed should increase linearly with n. The same linear scaling, but tending to a finite value as $N \to 1$ because of the finite speed of a single swimmer, is also found for a suspension of polar swimmers (Fig. 8.3(II)).

A measure of the nature of the collective motion generated by the interacting dumb-bells is the mean square displacement, $\langle \Delta r^2(t) \rangle_e = \langle |\mathbf{r}(t) - \mathbf{r}(0)|^2 \rangle_e$, where $\langle \cdot \rangle_e$ denotes an ensemble average. This is plotted in Fig. 8.3 for number densities of n = 0.02 and n = 0.1. At long times, and for large number densities, a scaling form $\langle \Delta r^2(t) \rangle_e \sim t^z$ develops, with an exponent of z = 1 indicating that the suspension of dumb-bells is behaving diffusively. At smaller number densities the behaviour is more sporadic because the large average separation between dumbbells greatly reduces the strength of the interactions between them. The collective motion is then dominated by those fluctuations in the local number density which bring two dumb-bells close enough to allow them to move appreciably.

The mean square displacement of a suspension of dumb-bells shows qualitatively different

behaviour to that of a suspension of polar swimmers. In the latter case, shown in Fig. 8.3(II), the mean square displacement is ballistic at short times (z = 2), with a cross-over to diffusion (z = 1) at longer times. Moreover, since this cross-over is due to the randomisation of swimmer orientations through hydrodynamic interactions, it occurs at later and later times as the number density is reduced. For polar swimmers this leads to a diffusion constant which decreases as the number density is increased. For apolar swimmers the opposite is true; their motion arises solely from hydrodynamic interactions and the diffusion constant increases with increasing number density.

8.6 Discussion

The aim of this Chapter has been to discuss the motion of a simple model of apolar swimmers, systems which move at zero Reynolds number *only* in the presence of other swimmers. We have demonstrated that, as is the case for colloids [179] and polar swimmers [165, 166], hydrodynamic interactions lead to complex collective behaviour.

Particular observations are that, for two dumb-bell swimmers which are initially parallel, the most likely final state corresponds to the axes of the swimmers lying at right angles. Oscillatory trajectories are also observed. For regular arrays of swimmers cooperation between hydrodynamic pair interactions can lead to simple flow fields. In particular, a square array of dumb-bells produces a constant flow and hence, if the dumb-bells were fixed in position, could act as a pump. For a large number of dumb-bell swimmers, initialised with random positions and phases, the mean speed is linear in the number of dumb-bells, reflecting the energy pumped into the system by the hydrodynamic interactions. The mean square displacement evolves linearly in time showing the expected diffusive behaviour.

There is much further work to be done to explore the phase space of apolar swimmers, for example considering initial configurations for which pairs move out of the plane, three dumbbell orbits, and the role of the distribution of phases on multi-dumb-bell motion. For more than two dumb-bells the relative phase becomes an important variable; since the pairwise interactions between three or more dumb-bells cannot simultaneously take their maximum value the system exhibits a type of frustration. It is also interesting to consider the effect of moving away from the zero Reynolds number limit [169], and to ask which of the properties of dumb-bells provide a generic representation of the class of apolar swimmers. Comparing simple microscopic models of polar and apolar swimming may help to formulate the correct continuum theory of swimmers, and to link the microscopic and continuum length scales.

CHAPTER 9

Conclusion

"What distressingly few of us realise is that almost all of these Taylors, except that of the Taylor series, are the <u>same</u> Taylor, Geoffrey Ingram."

The end of this thesis by no means marks the end of either of the two topics we have described. We take the opportunity to summarise our work and indicate some topics for future research.

Blue phases

Blue phases have seen a resurgence of interest in the past decade, prompted by the development of new experimental techniques to increase their stability and to exploit their unique properties for device applications. A thorough understanding of their response to an applied electric field is therefore important, since most liquid crystal devices rely upon the action of an electric field for their operation. Although this is a classic problem, worked on extensively in the 1980s, the agreement between theory and experiment has always been incomplete due to the inherent difficulty of the analytic calculations. In Chapter 3 we were able to show that the agreement may be improved, at least in part, by a numerical approach, which has the added benefit of describing both the static changes induced in the blue phase texture and the dynamics of their response.

Using a simple numerical technique for determining the redshift (§ 2.4) we were able to simulate the electrostriction of the cubic blue phases, obtaining for the first time both the correct order of magnitude of the components of the electrostriction tensor and the anomalous electrostriction of blue phase I (\S 3.1). The numerical approach allows for an unprecedented

description of the dynamics of electric field effects, allowing the pathway for textural transitions and complete unwinding of the blue phase to be determined. We were able to reproduce the blue phase I-blue phase X transition (§ 3.3) leading to the first determination of the detailed structure of the latter texture.

A full determination of the sequence of textural transitions that occur under the application of an electric field remains an important goal. Although a partial analytic calculation was performed by Hornreich *et al* in 1990 [79], the calculations were too extensive to allow blue phase X to be incorporated. The known differences between analytically and numerically calculated phase diagrams in zero field suggests that there may be similar discrepancies here. Furthermore, there is the interesting possibility that the three-dimensional hexagonal texture described by Hornreich *et al* [79] and included in their calculations may not be the correct phase. This is because they took their three-dimensional texture to be generated from the fundamental wavevectors for the two-dimensional hexagonal texture together with *higher* harmonics. However, there is also the possibility that the three-dimensional texture is generated by *sub*-harmonics, exactly as for the O^2 texture corresponding to blue phase II.

It would also be interesting to investigate experimentally the flexoelectric response of the blue phases [49]. Flexoelectric effects in the cholesteric phase offer a number of benefits for potential device applications, for example faster switching times and a response that is linear in the field strength for small fields, so they may afford similar benefits in blue phase devices. In Chapter 4 we identified three effects that might be expected for the blue phases: a change in the size and shape of the unit cell, or electrostriction, quadratic in the field strength, a distortion of the double twist cylinders at small fields, linear in the field strength, and a series of textural transitons to tetragonal and finally two-dimensional hexagonal textures. It is unfortunate that most of these effects are similar to those observed under dielectric coupling; the electrostriction is also quadratic in field strength and the textural transitions lead to structures with the same symmetries. This will undoubtedly make it difficult to distinguish between effects of dielectric and flexoelectric origin experimentally. The clearest indication that a phenomenon is flexoelectric in origin is if the response is linear in the field strength, as in the rotation of the optic axis in the flexoelectro-optic effect. The distortion of double twist cylinders in the blue phases represents one such linear response, which it may be possible to observe directly using confocal microscopy [84,85].

The origin of the stability of the wide temperature range blue phase discovered by Coles and Pivnenko [50] is still not understood. They have proposed that it might be flexoelectric in origin, based on the observation that the same bimesogenic molecules that yield the stable blue phase also exhibit large flexoelectric couplings. The status of polarisation fields produced internally in the cubic blue phases, and their consequences for energetic stability, remain to be determined.

Finally, a new direction that we are currently investigating is the influence of colloidal particles on the properties and stability of the blue phases. When a colloidal particle is added to a nematic liquid crystal, the anchoring conditions on the surface of the colloid are usually such that it is accompanied by the *creation* of disclination lines and a concomitant *increase* in the free energy. Not so for a blue phase, where the colloidal particles can attach themselves to the pre-existing disclination lines, occupying part of their position within the unit cell, and thus *removing* a length of disclination line. Since the disclination lines are energetically costly, if the energy associated with the surface anchoring of the colloid is not too large, this will lead to an overall *reduction* in the free energy, thereby aiding stabilisation of the blue phases.

Swimmers

Simple models, such as the Najafi-Golestanian swimmer, capture the essential feature of microorganisms in that they are able to swim by changing their shape. The usefulness of these models is that they remain analytically and numerically tractable without overly simplifying the details of their means of propulsion. The dependence of the hydrodynamic interactions on the properties of the swimming stroke can then be correctly determined and used as a guide to constructing more coarse-grained theories for the description of the collective behaviour of suspensions of swimmers.

We have shown in Chapter 6 that at a detailed, interswimmer level these interactions are not well described by the dipolar form suggested by simple scaling arguments and assumed in some previous studies [101,108,110]. Instead the interactions are higher order, decaying as r_{BA}^{-4} in the far field, and exhibit a sensitive dependence on the relative phase of the two swimmers. This sensitivity to the relative phase is important and allows for a fundamental distinction to be made between *active* and *passive* interactions.

An important next step is to extend the approach we developed in Chapter 6 to other simple models to see how substantially the details of the interactions vary between different already been performed, with a number of noteworthy features arising from the results. The most important is simply geometrical. Two vectors are required to describe the motion of a transverse swimmer: a vector \mathbf{n} giving the direction of its long axis and a vector \mathbf{m} giving the direction perpendicular to the long axis along which the oscillations take place.

Since, for such a swimmer, the forces act perpendicular to the long axis they will, in general, produce a torque that must be balanced by a bodily rotation so that the overall motion of the swimmer imparts no net torque on the surrounding fluid. We have found, however, that this bodily rotation can be eliminated by a suitable choice of the parameters of the swimming stroke, that choice corresponding precisely to the optimal swimming strategy. For this special case, the interactions between the swimmers both simplify greatly and exhibit some unexpected cancellations, leading to an active rotation that decays with separation as r_{BA}^{-5} and an active advection that decays even faster, as r_{BA}^{-6} .

The dependence of these active interactions on the relative phase is also interesting, appearing in both the advection and the rotation through a factor $\sin(\eta_{BA})$. Thus it is possible to eliminate the active interactions entirely by arranging for all the swimmers in a suspension to be exactly in phase, an option not available to Najafi-Golestanian swimmers since their active interactions vary cosinusoidally with the relative phase between them. Understanding the consequences this has for the collective behaviour of groups of the different types of swimmers will be an important future step.

Already it is clear that the nature of the swimming stroke of an individual can have an appreciable effect on the form of the detailed interactions between them. However, some features of swimmer-swimmer interactions are generic. For example, the scattering phenomena we described in Chapter 7 apply equally well to swimmers with transverse strokes, with the same types of scattering events, turn and exchange, and the exact recovery of the initial state, at least for planar scattering geometries. Indeed, our argument in Chapter 7 was that the phenomenon was due to symmetry, a consequence of kinematic reversibility, and therefore applicable to *any* swimmer, since, in principle, a T-dual can always either be found or fabricated.

Finally, we should like to be able to say more about the large scale behaviour of suspensions of swimmers and the appropriate form for a continuum description. Our analysis of apolar dumb-bells in Chapter 8 represents a start to this, but it is little more than a first step. Work is currently under way to extend these results and provide a more thorough comparison between the collective properties of polar and apolar swimmers. Our aim will be to make a connection to prominent features of continuum theories such as orientational ordering, spontaneous flow and anomalous density fluctuations.

References

- P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals*, 2nd ed. (Clarendon, Oxford, 1993).
- [2] I. W. Stewart, The Static and Dynamic Continuum Theory of Liquid Crystals: A Mathematical Introduction (Taylor & Francis, London, 2004).
- [3] P. M. Chaikin and T. C. Lubensky, *Principles of condensed matter physics* (Cambridge University Press, Cambridge, 1995).
- [4] S. Chandrasekhar, *Liquid Crystals*, 2nd ed., (Cambridge University Press, Cambridge, 1992).
- [5] M. J. Stephen and J. P. Straley, "Physics of liquid crystals", *Rev. Mod. Phys.* 46, 617 (1974).
- [6] I. Dierking, Textures of Liquid Crystals (Wiley-VCH, Weinheim, 2003).
- [7] R. B. Meyer, "Existence of even indexed disclinations in nematic liquid-crystals", *Phil. Mag.* 27, 405 (1973).
- [8] B. Chakrabarti, Y. Hatwalne, and N. V. Madhusudana, "Escape configuration lattice near the nematic-isotropic transition: Tilt analogue of blue phases", *Phys. Rev. Lett.* 96,157801 (2006).
- [9] G. P. Alexander and J. M. Yeomans, "Flexoelectric blue phases", *Phys. Rev. Lett.* 99, 067801 (2007).
- [10] N. Schopohl and T. J. Sluckin, "Defect core structure in nematic liquid crystals", *Phys. Rev. Lett.* 59, 2582 (1987).

- [11] D. C. Wright and N. D. Mermin, "Crystalline liquids: The blue phases", *Rev. Mod. Phys* 61, 385 (1989).
- [12] G. K. Batchelor, An introduction to fluid dynamics (Cambridge University Press, Cambridge, 1967).
- [13] L. M. Milne-Thomson, *Theoretical hydrodynamics* (Dover, New York, 1996).
- [14] J. Lighthill, Waves in fluids (Cambridge University Press, Cambridge, 1978).
- [15] F. M. Leslie, "Theory of flow phenomena in liquid crystals", in Advances in Liquid Crystals, Vol. 4, 1 (Academic Press, New York, 1979).
- [16] D. Forster, T. C. Lubensky, P. C. Martin, J. Swift, and P. S. Pershan, "Hydrodynamcis of liquid crystals", *Phys. Rev. Lett.* 26, 1016 (1971).
- [17] P. C. Martin, O. Parodi, and P. S. Pershan, "Unified hydrodynamic theory for crystals, liquid crystals and normal fluids", *Phys. Rev. A* 6, 2401 (1972).
- [18] G. Tóth, C. Denniston, and J. M. Yeomans, "Hydrodynamics of Topological Defects in Nematic Liquid Crystals", *Phys. Rev. Lett.* 88, 105504 (2002).
- [19] G. Tóth, C. Denniston, and J. M. Yeomans, "Hydrodynamics of domain growth in nematic liquid crystals", *Phys. Rev. E*, 67, 051705 (2003).
- [20] D. Svensek and S. Zumer, "Hydrodynamics of pair-annihilating disclination lines in nematic liquid crystals", *Phys. Rev. E* 66, 021712 (2002).
- [21] D. Svensek and S. Zumer, "Hydrodynamics of pair-annihilating disclinations in SmC films", Phys. Rev. Lett. 90, 155501 (2003).
- [22] P. Oswald and J. Ignés-Mullol, "Backflow-induced asymmetric collapse of disclination lines in liquid crystals", *Phys. Rev. Lett.* **95**, 027801 (2005).
- [23] C. Blanc, D. Svensek, S. Zumer, and M. Nobili, "Dynamics of nematic liquid crystal disclinations: The role of the backflow", *Phys. Rev. Lett.* **95**, 097802 (2005).
- [24] B. J. Edwards, A. N. Beris, and M. Grmela, "Generalized constitutive equation for polymeric liquid crystals: Part 1. Model formulation using the Hamiltonian (Poisson bracket) formulation", J. Non-Newtonian Fluid Mech. 35, 51 (1990).

- [25] B. J. Edwards, A. N. Beris, M. Grmela, and R. G. Larson, "Generalized constitutive equation for polymeric liquid crystals: Part 1. Non-homogeneous systems", J. Non-Newtonian Fluid Mech. 36, 243 (1990).
- [26] B. J. Edwards and A. N. Beris, "Non-canonical Poisson bracket for nonlinear elasticity with extensions to viscoelasticity", J. Phys. A: Math. Gen. 24, 2461 (1991).
- [27] B. J. Edwards, A. N. Beris. and M. Grmela, "The dynamic behavior of liquid-crystals a continuum description through generalized brackets", Mol. Cryst. Liq. Cryst. 201, 51 (1991).
- [28] A. N. Beris and B. J. Edwards, *Thermodynamics of Flowing Systems* (Oxford University Press, Oxford, 1994).
- [29] M. Grmela and H. C. Öttinger, "Dynamics and thermodynamics of complex fluids. I. Development of a general formalism", *Phys. Rev. E* 56, 6620 (1997).
- [30] H. C. Öttinger and M. Grmela, "Dynamics and thermodynamics of complex fluids. I. Illustrations of a general formalism", *Phys. Rev. E* 56, 6633 (1997).
- [31] H. C. Öttinger, Beyond Equilibrium Thermodynamics (WileyBlackwell, 2005)
- [32] C. Denniston, D. Marenduzzo, E. Orlandini, and J. M. Yeomans, "Lattice Boltzmann algorithm for three-dimensional liquid-crystal hydrodynamics", *Philos. Trans. R. Soc. London*, *Ser. A* 362, 1745 (2004).
- [33] J. M. Yeomans, "Mesoscale simulations: Lattice Boltzmann and particle algorithms", Physica A 369 159 (2006).
- [34] D. Marenduzzo, E. Orlandini, M. E. Cates, and J. M. Yeomans, "Steady-state hydrodynamic instabilities of active liquid crystals: Hybrid lattice Boltzmann simulations", *Phys. Rev. E* 76, 031921 (2007).
- [35] P. Pieranski, R. Barbet-Massin, and P. E. Cladis, "Steps on surfaces of liquid-crystal blue phase I", Phys. Rev. A 31, 3912 (1985).
- [36] S. Meiboom, J. P. Sethna, P. W. Anderson, and W. F. Brinkman, "Theory of the blue phase of cholesteric liquid crystals", *Phys. Rev. Lett.* 46, 1216 (1981).

- [37] S. Meiboom, M. Sammon, and W. F. Brinkman, "Lattice of disclinations: The structure of the blue phases of cholesteric liquid crystals", *Phys. Rev. A* 27, 438 (1983).
- [38] R. M. Hornreich, M. Kugler, and S. Shtrikman, "Localized instabilities and the orderdisorder transition in cholesteric liquid crystals", *Phys. Rev. Lett.* 48, 1404 (1982).
- [39] T. Blümel, P. J. Collings, H. Onusseit, and H. Stegemeyer, "Phase diagrams of the blue phases", *Chem. Phys. Lett.* **116**, 529 (1985).
- [40] D. K. Yang and P. P. Crooker, "Chiral-racemic phase diagrams of blue-phase liquid crystals", Phys. Rev. A 35, 4419 (1987).
- [41] R. M. Hornreich and S. Shtrikman, "Theory of structure and properties of cholesteric blue phases", Phys. Rev. A 24, 635 (1981).
- [42] H. Grebel, R. M. Hornreich, and S. Shtrikman, "Landau theory of cholesteric blue phases", *Phys. Rev. A* 28, 1114 (1983).
- [43] H. Grebel, R. M. Hornreich, and S. Shtrikman, "Landau theory of cholesteric blue phases - the role of higher haarmonics", *Phys. Rev. A* 30, 3264 (1984).
- [44] T. C. Lubensky and H. Stark, "Theory of a critical point in the blue phase III-isotropic phase diagram", Phys. Rev. E 53, 714 (1996).
- [45] M. A. Anisimov, V. A. Agayan, and P. J. Collings, "Nature of the blue phase III-isotropic critical point: An analogy with the liquid gas transition", *Phys. Rev. E* 57, 582 (1998).
- [46] W. Cao, A. Muñoz, P. Palffy-Muhoray, and B. Taheri, "Lasing in a three-dimensional photonic crystal of the liquid crystal blue phase II", *Nat. Mater.* 1, 111 (2002).
- [47] Y. Hisakado, H. Kikuchi, T. Nagamura, and T. Kajiyama, "Large electro-optic Kerr effect in polymer-stabilized liquid-crystalline blue phases", Adv. Mater. 17, 96 (2005).
- [48] S. Yokoyama, S. Mashiko, H. Kikuchi, K. Uchida, and T. Nagamura, "Laser emission from a polymer-stabilized liquid-crystalline blue phase", Adv. Mater. 18, 48 (2006).
- [49] H.-S. Kitzerow, "Blue phases at work!", ChemPhysChem 7, 63 (2006).
- [50] H. J. Coles and M. N. Pivnenko, "Liquid crystal 'blue phases' with a wide temperature range", *Nature* 436, 997 (2005).

- [51] H. Kikuchi, M. Yokota, Y. Hisakado, H. Yang, and T. Kajiyama, "Polymer-stabilized liquid crystal blue phases", *Nat. Mater.* 1, 64 (2002).
- [52] http://en.wikipedia.org/wiki/Blue_Phase_Mode_LCD, retrieved 11th June 2008.
- [53] H.-S. Kitzerow, "The effect of electric fields on blue phases", Mol. Cryst. Liq. Cryst. 202, 51 (1991).
- [54] A. Dupuis, D. Marenduzzo, and J. M. Yeomans, "Numerical calculation of the phase diagram of the cubic blue phases in cholesteric liquid crystals", *Phys. Rev. E* 71, 011703 (2005).
- [55] J. M. Ball, Graduate lecture course on liquid crystals, University of Oxford (Trinity Term 2006).
- [56] D. W. Berreman and S. Meiboom, "Tensor representation of Oseen-Frank strain energy in uniaxial cholesterics", *Phys. Rev. A* 30, 1955 (1984).
- [57] L. Longa and H.-R. Trebin, "Structure of the elastic free energy for chiral nematic liquid crystals", *Phys. Rev. A* 39, 2160 (1989).
- [58] L. Longa, D. Monselesan, and H.-R. Trebin, "An extension of the Landau-Ginzburg-de Gennes theory for liquid-crystals", *Liq. Cryst.* 2, 769 (1987).
- [59] J. Jung, C. Denniston, E. Orlandini, and J. M. Yeomans, "Anisotropy of domain growth in nematic liquid crystals", *Liq. Cryst.* **30**, 1455 (2003).
- [60] S. A. Brazovskii and S. G. Dmitriev, "Phase-transitions in cholesteric liquid-crystals", Zh. Eksp. Teor. Fiz. 69, 979 (1975) [Sov. Phys. JETP 42, 497 (1975)].
- [61] V. A. Belyakov and V. E. Dmitrienko, "Blue phase of liquid-crystals", Usp. Fiz. Nauk
 146, 369 (1985) [Sov. Phys. Usp. 28, 535 (1985)].
- [62] B. P. Huff, J. J. Krich and P. J. Collings, "Helix inversion in the chiral nematic and isotropic phases of a liquid crystal", *Phys. Rev. E* 61, 5372 (2000).
- [63] A. J. Slaney, I. Nishiyama, P. Styring, and J. W. Goodby, "Twist inversion in a cholesteric material containing a single chiral center", J. Mater. Chem 2 (8), 805 (1992).

- [64] M. J. Buerger, "Crystallographic symmetry in reciprocal space", Proc. Natl. Acad. Sci. U.S. 35, 198 (1949).
- [65] L. Longa, D. Monselesan, and H.-R. Trebin, "Phase-diagrams of cholesteric liquid-crystals obtained with a generalized Landau-de Gennes theory", *Liq. Cryst.* 5, 889 (1989).
- [66] R. J. Miller and H. F. Gleeson, "The influence of pretransitional phenomena on blue phase range", *Liq. Cryst.* 14, 2001 (1993).
- [67] H. Stegemeyer and F. Porsch, "Electric field effect on phase transitions in liquid-crystalline blue-phase systems", *Phys. Rev. A* 30, 3369 (1984).
- [68] F. Porsch and H. Stegemeyer, "The effect of an electric field on the selective reflection bands of liquid crystalline blue phases", *Chem. Phys. Lett.* **125**, 319 (1986).
- [69] F. Porsch and H. Stegemeyer, "Electrostriction of liquid crystalline blue phases", Chem. Phys. Lett. 155, 620 (1989).
- [70] P. Pieranski, P. E. Cladis, and R. Barbet-Massin, "Experimental evidence for a hexagonal blue phase", J. Physique Lett. 46, L-973 (1985).
- [71] P. E. Cladis, T. Garel. and P. Pieranski, "Kossel diagrams show electric-field-induced cubic-tetragonal structural transition in frustrated liquid-crystal blue phases", *Phys. Rev. Lett.* 57, 2841 (1986).
- [72] P. Pieranski and P. E. Cladis, "Field induced tetragonal blue phase (BP-X)", *Phys. Rev.* A 35, 355 (1987).
- [73] M. Jorand and P. Pieranski, "Brownian motion of screw dislocations in the hexagonal blue phase", J. Physique 48, 1197 (1987).
- [74] G. Heppke, B. Jerome, H.-S. Kitzerow, and P. Pieranski, "Observation of a hexagonal blue phase in systems with negative dielectric anisotropy", *Liq. Cryst.* 5, 813 (1989).
- [75] N. Chen and J. Ho, "Electric-field-induced phase diagrams of blue-phase systems", Phys. Rev. A 35, 4886 (1987).
- [76] R. M. Hornreich, M. Kugler, and S. Shtrikman, "Possibility of a field-induced hexagonal blue phase in cholesteric liquid crystals", *Phys. Rev. Lett.* 54, 2099 (1985).

- [77] D. Lubin and R. M. Hornreich, "Effect of a weak electric field on the cubic blue phases of cholesteric liquid crystals", *Phys. Rev. A* 36, 849 (1987).
- [78] H. Stark and H.-R. Trebin, "Theory of electrostriction of liquid-crystalline blue phase I and II", Phys. Rev. A 44, 2752 (1991).
- [79] R. M. Hornreich and S. Shtrikman, "Field-induced hexagonal blue phases in positive and negative dielectric anisotropy systems: Phase diagrams and topological properties", *Phys. Rev. A* 41, 1978 (1990).
- [80] L. Longa, M. Zelazna, H.-R. Trebin, and J. Mościcki, "Electrostriction of the cubic blue phases in the presence of bond orientational order", *Phys. Rev. E* 53, 6067 (1996).
- [81] M. Zelazna, L. Longa, H.-R. Trebin, and H. Stark, "Electrostriction of the cubic blue phases: The role of amplitudes", *Phys. Rev. E* 57, 6711 (1998).
- [82] G. P. Alexander and J. M. Yeomans, "Stabilizing the blue phases", Phys. Rev. E 74, 061706 (2006).
- [83] A. Dupuis, D. Marenduzzo, E. Orlandini, and J. M. Yeomans, "Rheology of cholesteric blue phases", *Phys. Rev. Lett.* 95, 097801 (2005).
- [84] H. Kikuchi, S. Hirata, and K. Uchida, "Confocal laser scanning microscopic observation of polymer-stabilized blue phase I", Mol. Cryst. Liq. Cryst., 465, 283 (2007).
- [85] K. Higashiguchi, K. Yasui, and H. Kikuchi, "Direct observation of polymer-stabilized blue phase I structure with confocal laser scanning microscope", J. Am. Chem. Soc. 130, 6326 (2008).
- [86] International Tables for Crystallography, 5th ed., Editor Th. Hahn (Springer, 2005).
- [87] R. B. Meyer, "Piezoelectric effects in liquid crystals", Phys. Rev. Lett. 22, 918, (1969).
- [88] J. Prost and P. S. Pershan, "Flexoelectricity in nematic and smectic A liquid-crystals", J. Appl. Phys. 47, 2298 (1976).
- [89] G. P. Bryan-Brown, C. V. Brown, E. L. Wood, I. C. Sage, P. Brett, and J. Rudin, SID Int. Symp. Digest Tech. Papers 28, 37 (1997).

- [90] C. Denniston and J. M. Yeomans, "Flexoelectric surface switching of bistable nematic devices", *Phys. Rev. Lett.* 87, 275505 (2001).
- [91] A. J. Davidson and N. J. Mottram, "Flexoelectric switching in a bistable nematic device", *Phys. Rev. E* 65, 051710 (2002).
- [92] C. Uche, S. J. Elston, and L. A. Parry-Jones, "Microscopic observation of zenithal bistable switching in nematic devices with different surface relief structures", J. Phys. D: Appl. Phys. 38, 2283 (2005).
- [93] L. A. Parry-Jones and S. J. Elston, "Flexoelectric switching in a zenithally bistable nematic device", J. Appl. Phys. 97, 093515 (2005).
- [94] J. S. Patel and R. B. Meyer, "Flexoelectric electro-optics of a cholesteric liquid crystal", *Phys. Rev. Lett.* 58, 1538 (1987).
- [95] H. J. Coles, M. J. Clarke, S. M. Morris, B. J. Broughton, and A. E. Blatch, "Strong flexoelectric behavior in bimesogenic liquid crystals", J. Appl. Phys. 99, 034104 (2006).
- [96] P. Rudquist, M. Buivydas, L. Komitov, and S. T. Lagerwall, "Linear electro-optic effect based on flexoelectricity in a cholesteric with sign change of dielectric anisotropy", J. Appl. Phys. 76, 7778 (1994).
- [97] B. J. Broughton, M. J. Clarke, A. E. Blatch, and H. J. Coles, "Optimized flexoelectric response in a chiral liquid-crystal phase device", J. Appl. Phys. 98, 034109 (2005).
- [98] J. Toner, Y. Tu, and S. Ramaswamy, "Hydrodynamics and phases of flocks", Ann. Phys. 318, 170 (2005).
- [99] S. Ramaswamy and R. A. Simha, "The mechanics of active matter: Broken-symmetry hydrodynamics of motile particles and granular layers", *Solid State Commun.* 139, 617 (2006).
- [100] T. Vicsek, A. Czirok, E. Ben-Jacob, I. Cohen, and O. Shochet, "Novel type of phase transition in a system of self-driven particles", *Phys. Rev. Lett.* **75**, 1226 (1995).
- [101] T. J. Pedley and J. O. Kessler, "Hydrodynamic phenomena in suspensions of swimming microorganisms", Ann. Rev. Fluid Mech. 24, 313 (1992).

- [102] X-L. Wu and A. Libchaber, "Particle diffusion in a quasi-two-dimensional bacterial bath", *Phys. Rev. Lett.* 84, 3017 (2000).
- [103] C. Dombrowski, L. Cisneros, S. Chatkaew, R. E. Goldstein, and J. O. Kessler, "Selfconcentration and large-scale coherence in bacterial dynamics", *Phys. Rev. Lett.* 93, 098103 (2004).
- [104] H. C. Berg, "Motile behavior of bacteria", *Physics Today* 53, 24 (2000).
- [105] E. O. Budrene and H. C. Berg, "Complex patterns formed by motile cells of *Escherichia coli*", *Nature* 349, 630 (1991).
- [106] E. O. Budrene and H. C. Berg, "Dynamics of formation of symmetrical patterns by chemotactic bacteria", *Nature* 376, 49 (1995).
- [107] L. Cisneros, C. Dombrowski, R. E. Goldstein, and J. O. Kessler, "Reversal of bacterial locomotion at an obstacle", *Phys. Rev. E* 73, 030901(R) (2006).
- [108] R. A. Simha and S. Ramaswamy, "Hydrodynamic fluctuations and instabilities in ordered suspensions of self-propelled particles", *Phys. Rev. Lett.* 89, 058101 (2002).
- [109] Y. Hatwalne, S. Ramaswamy, M. Rao, and R. A. Simha, "Rheology of active-particle suspensions", *Phys. Rev. Lett.* **92**, 118101 (2004).
- [110] J. P. Hernandez-Ortiz, C. G. Stoltz, and M. D. Graham, "Transport and collective dynamics in suspensions of confined swimming particles", *Phys. Rev. Lett.* 95, 204501 (2005).
- [111] I. Llopis and I. Pagonabarraga, "Dynamic regimes of hydrodynamically coupled selfpropelling particles", *Europhys. Lett.* 75, 999 (2006).
- [112] L. H. Cisneros, R. Cortez, C. Dombrowski, R. E. Goldstein, and J. O. Kessler, "Fluid dynamics of self-propelled micro-organisms, from individuals to concentrated populations", *Expt. in Fluids* 43, 737 (2007).
- [113] D. Saintillan and M. J. Shelley, "Orientational order and instabilities in suspensions of self-locomoting rods", *Phys. Rev. Lett.* **99**, 058102 (2007).
- [114] G. I. Taylor, "Analysis of the swimming of microscopic organisms", Proc. R. Soc. London A 209, 447 (1951).

- [115] G. I. Taylor, "The action of waving cylindrical tails in propelling microscopic organisms", Proc. R. Soc. London A 211, 225 (1952).
- [116] G. I. Taylor, Low Reynolds Number Flows (Encyclopaedia Britannica Educational Corp., Chicago), Video No. 21617 (1967).
- [117] E. M. Purcell, "Life at low Reynolds number", Am. J. Phys. 45, 3 (1977).
- [118] A. Shapere and F. Wilczek, "Self-propulsion at low Reynolds number", *Phys. Rev. Lett.* 58, 2051 (1987).
- [119] A. Shapere and F. Wilczek, "Geometry of self-propulsion at low Reynolds number", J. Fluid Mech. 198, 557 (1989).
- [120] A. Shapere and F. Wilczek, "Efficiencies of self-propulsion at low Reynolds number", J. Fluid Mech. 198, 587 (1989).
- [121] J. Lighthill, "Flagellar hydrodynamics: The John von Neumann Lecture, 1975", SIAM Review, 18 (2), 161 (1976).
- [122] H. A. Stone and A. D. T. Samuel, "Propulsion of microorganisms by surface distortions", *Phys. Rev. Lett.* 77, 4102 (1996).
- [123] C. H. Wiggins and R. E. Goldstein, "Flexive and propulsive dynamics of elastica at low Reynolds number", *Phys. Rev. Lett.* 80, 3879 (1998).
- [124] D. L. Hu, B. Chan, and J. W. M. Bush, "The hydrodynamics of water strider locomotion", *Nature* 424, 663 (2003).
- [125] D. L. Hu and J. W. M. Bush, "Meniscus-climbing insects", Nature 437, 733 (2005).
- [126] D. Tam and A. E. Hosoi, "Optimal stroke patterns for Purcell's three-link swimmer", *Phys. Rev. Lett.* 98, 068105 (2007).
- [127] H. C. Berg and R. A. Anderson, "Bacteria swim by rotating their flagellar filaments", *Nature (London)* 245, 380 (1973).
- [128] L. Turner, W. S. Ryu, and H. C. Berg, "Real time imaging of fluorescent flagellar filaments", J. Bacteriol. 182, 2793 (2000).

- [129] J. J. L. Higdon, "The generation of feeding currents by flagellar motions", J. Fluid Mech. 94, 305 (1979).
- [130] J. J. L. Higdon, "The hydrodynamics of flagellar propulsion: Helical waves", J. Fluid Mech. 94, 331 (1979).
- [131] R. E. Goldstein, T. R. Powers, and C. H. Wiggins, "Viscous nonlinear dynamics of twist and writhe", *Phys. Rev. Lett.* 80, 5232 (1998).
- [132] R. E. Goldstein, A. Goriely, G. Huber, and C. W. Wolgemuth, "Bistable helices", Phys. Rev. Lett. 84, 1631 (2000).
- [133] D. Coombs, G. Huber, J. O. Kessler, and R. E. Goldstein, "Periodic chirality transformations propagating on bacterial flagella", *Phys. Rev. Lett.* 89, 118102 (2002).
- [134] J. W. Shaevitz, J. Y. Lee, and D. A. Fletcher, "Spiroplasma swim by a processive change in body helicity", *Cell* **122**, 941 (2005).
- [135] H. Wada and R. Netz, "Model for self-propulsive helical filaments: Kink-pair propagation", Phys. Rev. Lett. 99, 108102 (2007).
- [136] L. E. Becker, S. A. Koehler, and H. A. Stone, "On self-propulsion of micro-machines at low Reynolds number: Purcell's three-link swimmer", J. Fluid Mech. 490, 15 (2003).
- [137] R. Dreyfus, J. Baudry, and H. A. Stone, "Purcell's "rotator": Mechanical rotation at low Reynolds number", *Eur. Phys. J. B* 47, 161 (2005).
- [138] I. M. Kulic, R. Thaokar, and H. Schiessel, "Twirling DNA rings swimming nanomotors ready for a kickstart", *Europhys. Lett.* 72, 527 (2005).
- [139] R. M. Thaokar, H. Schiessel, and I. M. Kulic, "Hydrodynamics of a rotating torus", Eur. Phys. J. B 60, 325 (2007).
- [140] A. Najafi and R. Golestanian, "Simple swimmer at low Reynolds number: Three linked spheres", Phys. Rev. E 69, 062901 (2004).
- [141] R. Golestanian and A. Ajdari, "Mechanical response of a small swimmer driven by conformational transitions", *Phys. Rev. Lett.* 100, 038101 (2008).
- [142] R. Golestanian, "Three-sphere low-Reynolds-number swimmer with a cargo container", Eur. Phys. J. E 25, 1 (2008).
- [143] R. Golestanian and A. Ajdari, "Analytic results for the three-sphere swimmer at low Reynolds number", Phys. Rev. E 77, 036308 (2008).
- [144] J. E. Avron, O. Kenneth, and D. H. Oaknin, "Pushmepullyou: An efficient microswimmer", New J. Phys. 7, 234 (2005).
- [145] D. J. Earl, C. M. Pooley, J. F. Ryder, I. Bredburg, and J. M. Yeomans, "Modelling microscopic swimmers at low Reynolds number", J. Chem. Phys. 126, 064703 (2007).
- [146] R. Golestanian, T. B. Liverpool, and A. Ajdari, "Propulsion of a molecular machine by asymmetric distribution of reaction products", *Phys. Rev. Lett.* 94, 220801 (2005).
- [147] R. Golestanian, T. B. Liverpool, and A. Ajdari, "Designing phoretic micro- and nanoswimmers", New J. Phys. 9, 126 (2007).
- [148] R. Dreyfus, J. Baudry, M. L. Roper, M. Fermigier, H. A. Stone, and J. Bibette, "Microscopic artificial swimmers", *Nature* 437, 862 (2005).
- [149] M. Roper, R. Drefus, J. Baudry, M. Fermigier, J. Bibette, and H. A. Stone, "On the dynamics of magnetically driven elastic filaments", J. Fluid. Mech. 554, 167 (2006).
- [150] J. R. Howse, R. A. L. Jones, A. J. Ryan, T. Gough, R. Vafabakhsh, and R. Golestanian, "Self-motile colloidal particles: From directed propulsion to random walk", *Phys. Rev. Lett.* 99, 048102 (2007).
- [151] J. O. Kessler, "Hydrodynamic focusing of motile algal cells", Nature 313, 218 (1985).
- [152] D. C. Guell, H. Brenner, R. B. Frankell, and H. Hartman, "Hydrodynamic forces and band formation in swimming magnetotactic bacteria", J. Theor. Biol. 135, 525 (1988).
- [153] D. C. Guell, "Hydrodynamic forces in bands of magnetotactic bacteria, revisited: Formation, stability and structure", *Chem. Eng. Commun.* 150, 59 (1996).
- [154] I. H. Riedel, K. Kruse, and J. Howard, "A self-organized vortex array of hydrodynamically entrained sperm cells", *Science* **309**, 300 (2005).

- [155] W. R. DiLuzio, L. Turner, M. Mayer, P. Garstecki, D. B. Weibel, H. C. Berg, and G. M. Whitesides, "Escherichia coli swim on the right-hand side", Nature 435, 1271 (2005).
- [156] E. Lauga, W. R. DiLuzio, G. M. Whitesides, and H. A. Stone, "Swimming in circles: Motion of bacteria near solid boundaries", *Biophys. J.* 90, 400 (2006).
- [157] J. Hill, O. Kalkanci, J. L. McMurry, and H. Koser, "Hydrodynamic surface interactions enable *Escerichia coli* to seek efficient routes to swim upstream", *Phys. Rev. Lett.* 98, 068101 (2007).
- [158] J. Lighthill, "Helical distributions of stokeslets", J. Eng. Math. 30: 35 (1996).
- [159] M. Ramia, D. L. Tullock, and N. Phan-Thien, "The role of hydrodynamic interaction in the locomotion of microorganisms", *Biophys. J.* 65, 755 (1993).
- [160] S. Nasseri and N. Phan-Thien, "Hydrodynamic interaction between two nearby swimming micromachines", J. Comp. Mech. 20, 551 (1997).
- [161] M. Kim and T. R. Powers, "Hydrodynamic interactions between rotating helices", Phys. Rev. E 69, 061910 (2004).
- [162] T. Ishikawa, M. P. Simmonds, and T. J. Pedley, "Hydrodynamic interaction of two swimming model micro-organisms", J. Fluid Mech. 568, 119 (2006).
- [163] T. Ishikawa and M. Hota, "Interaction of two swimming Paramecia", J. Exp. Biol. 209, 4452 (2006).
- [164] T. Ishikawa, G. Sekiya, Y. Imai, and T. Yamaguchi, "Hydrodynamic interactions between two swimming bacteria", *Biophys. J* 93, 2217 (2007).
- [165] T. Ishikawa and T. J. Pedley, "Coherent structures in monolayers of swimming particles", *Phys. Rev. Lett.* **100**, 088103 (2008).
- [166] C. M. Pooley, G. P. Alexander, and J. M. Yeomans, "Hydrodynamic interaction between two swimmers at low Reynolds number", *Phys. Rev. Lett.* 99, 228103 (2007).
- [167] E. E. Keavney and M. R. Maxey, "Interactions between comoving magnetic microswimmers", Phys. Rev. E 77, 041910 (2008).

- [168] R. M. Thaokar, "Hydrodynamic interaction between two rotating tori", *Eur. Phys. J. B* 61, 47 (2008).
- [169] E. Lauga, "Continuous breakdown of Purcell's scallop theorem with inertia", *Phys. Fluids* 19, 061703 (2007).
- [170] J. Happel and H. Brenner, Low Reynolds Number Hydrodynamics (Prentice-Hall, Englewood Cliffs, NJ, 1965).
- [171] J. R. Blake and A. T. Chwang, "Fundamental singularities of viscous flow. Part I: The image systems in the vicinity of a stationary no-slip boundary", J. Eng. Math. 8, 23 (1974).
- [172] J. Koiller, K. Ehlers, and R. Montgomery, "Problems and progress in microswimming", J. Nonlinear Sci. 6, 507 (1996).
- [173] V. Narayan, S. Ramaswamy, and N. Menon, "Long-lived giant number fluctuations in a swarming granular nematic", *Science* **317**, 105 (2007).
- [174] H. Chaté, F. Ginelli, and R. Montagne, "Simple model for active nematics: Quasi-longrange order and giant fluctuations", *Phys. Rev. Lett.* 96, 180602 (2006).
- [175] D. Marenduzzo, E. Orlandini, and J. M. Yeomans, "Hydrodynamics and rheology of active liquid crystals: A numerical investigation", *Phys. Rev. Lett.* 98, 118102 (2007).
- [176] R. Voituriez, J. F. Joanny, and J. Prost, "Spontaneous flow transition in active polar gels", *Europhys. Lett.* 70, 404 (2005).
- [177] K. Kruse, J. F. Joanny, F. Jülicher, J. Prost, and K. Sekimoto, "Generic theory of active polar gels: A paradigm for cytoskeletal dynamics", *Eur. Phys. J. E* 16, 5 (2005).
- [178] R. Voituriez, J. F. Joanny, and J. Prost, "Generic phase diagram of active polar films", *Phys. Rev. Lett.* 96, 028102 (2006).
- [179] I. M. Jánosi, T. Tél, D. E. Wolf, and J. A. C. Gallas, "Chaotic particle dynamics in viscous flows: The three-particle stokeslet problem", *Phys. Rev. E* 56, 2858 (1997).