

## SUMMARY OF PROFESSIONAL ACCOMPLISHMENTS

### 1 NAME

**Jeffrey Christopher Everts**

### 2 DIPLOMAS, DEGREES CONFERRED IN SPECIFIC AREAS OF SCIENCE OR ARTS

- **Ph.D. degree in Theoretical Physics**, 19 September 2016  
Institute for Theoretical Physics, Utrecht University, the Netherlands  
Thesis: *Colloidal dispersions of repulsive nanoparticles: tunable effective interactions, phase behaviour and anisotropy.*  
Supervisor: prof. dr. René van Roij
- **M.Sc. degree in Theoretical Physics (cum laude)**, 27 August 2012  
Utrecht University, the Netherlands  
Thesis: *Topological phases and fermionic superfluidity on the Lieb lattice*  
Supervisor: prof. dr. Cristiane de Morais Smith
- **B.Sc. degree in Applied Physics (cum laude)**, 25 August 2010  
Eindhoven University of Technology, the Netherlands  
Thesis: *Expression of chirality by competing nucleated self-assembly*  
Supervisor: prof. dr. ir. Paul van der Schoot
- **B.Sc. degree in Chemical Engineering and Chemistry (cum laude)**, 25 August 2010  
Eindhoven University of Technology, the Netherlands  
Thesis: *Expression of chirality in deuterated benzene-1,3,5, tricarboxamides*  
Supervisor: dr. Anja Palmans

### 3 INFORMATION ON EMPLOYMENT IN RESEARCH INSTITUTES OR FACULTIES/DEPARTMENTS

- **Assistant Professor**, October 2022 – present,  
Institute of Theoretical Physics, Faculty of Physics, University of Warsaw, Poland.
- **Postdoctoral Research Associate**, July 2020 – present,  
Institute of Physical Chemistry, Polish Academy of Sciences, Warsaw, Poland.
- **Visiting Scientist**, April 2019 – May 2019,  
University of Cambridge, Cambridge, United Kingdom.
- **Postdoctoral Research Associate**, January 2017 – July 2020,  
Faculty for Mathematics and Physics, University of Ljubljana, Slovenia.
- **Ph.D. Student**, September 2012 – September 2016,  
Institute for Theoretical Physics, Utrecht University, the Netherlands.
- **Junior researcher**, April 2008 – October 2010,  
Spinoza Institute, Eindhoven University of Technology, the Netherlands.

## 4 DESCRIPTION OF THE ACHIEVEMENTS, SET OUT IN ART. 219 PARA 1 POINT 2 OF THE ACT

### A. TITLE OF THE SCIENTIFIC ACHIEVEMENT

A series of scientific articles related thematically entitled  
*Electric double layers in anisotropic fluids*

### B. LIST OF PUBLICATIONS FORMING THE SCIENTIFIC ACHIEVEMENT (reverse chronological order) (†corresponding author, \*equal contribution)

- [A1] C. Dao, J. C. Everts, M. Ravnik, and Y. Tserkovnyak,  
*Nematronics: Reciprocal coupling between ionic currents and nematic dynamics*,  
Phys. Rev. Lett. **132**, 218303 (2023).
- [A2] J. C. Everts<sup>†</sup> and M. Ravnik,  
*Ionically charged topological defects in nematic fluids*,  
Phys. Rev. X **11**, 011054 (2021).
- [A3] J. C. Everts<sup>\*</sup>, B. Senyuk<sup>\*</sup>, H. Munder, M. Ravnik and I. I. Smalyukh,  
*Anisotropic electrostatic screening of charged colloids in nematic solvents*,  
Sci. Adv. **7**, eabd0662 (2021).
- [A4] J. C. Everts<sup>†</sup> and M. Ravnik,  
*Charge-, salt- and flexoelectricity-driven anchoring effects in nematics*,  
Liq. Cryst. **48**, 423 (2021).
- [A5] M. Ravnik and J. C. Everts<sup>†</sup>,  
*Topological-defect-induced surface charge heterogeneities in nematic electrolytes*,  
Phys. Rev. Lett. **125**, 037801 (2020).

*Detailed information about my contribution to the articles above is collected in Załącznik nr 4 (Appendix 4).*

### C. DESCRIPTION OF THE SCIENTIFIC ACHIEVEMENT

#### 1. Introduction

Liquid crystals are phases of matter which, due to their long-ranged ordered macroscopic structure, exhibit strongly anisotropic physical properties [1]. In some cases, liquid crystals flow like an ordinary liquid [2–4], whereas in other cases, they deform elastically as a crystalline solid [5, 6]. From a symmetry-breaking point of view, such properties can be understood from their partially ordered structure: the rotational symmetry of an isotropic liquid is broken by a liquid crystal, but the translational symmetry stays intact or is only partially broken. Clearly, this is only possible when the interactions between the constituent particles of a liquid crystal (the so-called mesogens) are anisotropic themselves. It is not a surprise that for the many possibilities of realising such anisotropic interactions (e.g., by the particle shape [7, 8] or by utilizing specific chemical groups [9]), one can realise various types of liquid crystals. The simplest type of liquid crystals are characterised by pure nematic order. For such systems, the mesogens (e.g., rods) all point on average in the same direction, but

their center-of-masses do not exhibit any long-range translational order. Furthermore, there is a residual up-down  $\mathbb{Z}_2$  symmetry, which makes such systems different from polar phases like ferromagnets. Nematic liquid crystals are realised in various systems. Examples include rod-like molecular systems (e.g. 4-cyano-4'-pentylbiphenyl, typically abbreviated as 5CB [10]), but also colloidal systems with rod-shaped particles, like the tobacco mosaic virus [11] or bacterial colonies [12].

In practice, nematic liquid crystals are rarely (pure) one-component systems. Colloidal nematics, for example, are immersed in a solvent (e.g., water), and molecular liquid crystals often have many impurities or dopants. Common to both types of liquid crystals is that the particles forming the nematic phase are often accompanied by ions [13–15]. Despite their abundance in liquid crystals, the role of ions in these systems is, surprisingly, poorly understood, contrasting their well-studied status in isotropic liquids. In isotropic liquids, it is well known that ions –when brought into contact with charged surfaces – form structures that are known as electric double layers [16]. An electric double layer consists of two layers of charge: (i) the charge distribution on the surface and (ii) a diffuse charge layer residing in the liquid that is in contact with the charged surface. This latter diffuse layer is formed due to the competition of entropy, favouring a homogeneous ion distribution, and the Coulomb interaction, which, e.g., favours that ions are in a bound state [17] or that ions of the opposite charge are as close as possible to the charged surface. In this achievement, we describe the spatial structure of such electric double layers in nematic liquid crystals, and in particular, we showcase some of the consequences of their presence.

The papers [A1-A5] selected for the scientific achievement describe important contributions to understanding the effects of electric double layers in nematic liquid crystals, in and out of equilibrium. In our work, we systematically increased the complexity of the systems to understand the various effects resulting from the cross-talk of ions interacting with a nematic background. In addition, these results also lead to new questions, which open many doors for novel theoretical and experimental studies. Here, we present the results on

- (i) The effect of screened electric fields on the orientation of mesogens close to a charged surface [A4].
- (ii) Anisotropic screening and its effects on the inter-particle interactions of charged colloidal particles in a nematic medium [A3].
- (iii) The structure of electric double layers around topological defects and how they affect charged surfaces [A2, A5].
- (iv) The reciprocal effects between liquid-crystal dynamics and ion electrokinetics, including the consequences in some simple geometries [A1].

The structure of the description is as follows. To have a self-contained presentation of the results, we first spend some time on the type of models employed in these studies in Sec. C.2. Then, we present the main results of the scientific achievement in Sec. C.3. In Sec. C.4, we conclude with a summary and a perspective on how these results will guide future research in this field, including my future plans.

## 2. Mesoscopic modelling of electrostatic and electrokinetic effects in nematic fluids

### I. Equilibrium effects

Describing a mixture of anisotropic particles with ions from a microscopic point of view is a challenging task. It is, therefore, unavoidable to resort to approximations. Furthermore, starting with the simplest possible models is always beneficial to obtain indispensable insights. Since ions are well-studied

in isotropic liquids, we have an important source of inspiration for describing ion-doped nematics. For simplicity, we take just two species of monovalent ions consisting of cations (+) and anions (-), described by their (local) number density profiles  $\rho_{\pm}(\mathbf{r})$ . Inspired by the well-known Poisson-Boltzmann theory for electric double layers in ordinary liquids [18], we assume that the dielectric properties of the nematic medium are described by the dielectric tensor  $\epsilon$ . Unlike isotropic liquids, a nematic medium cannot be taken to be structureless. In this case, we describe the nematic order by the tensorial (quadrupolar) order parameter  $\mathbf{Q}(\mathbf{r})$ . Here, the largest eigenvalue of this symmetric and traceless rank-2 tensor, known as the scalar order parameter  $S(\mathbf{r})$ , describes the degree of orientational order along the accompanied (normalised) eigenvector  $\hat{\mathbf{n}}$ , defined as the nematic director. The director is a measure for the average orientation of the mesogens and satisfies the equivalence relation  $\hat{\mathbf{n}} \sim -\hat{\mathbf{n}}$  due to the aforementioned up-down symmetry. With the help of  $\mathbf{Q}(\mathbf{r})$  we can parametrize the dielectric tensor as [1]

$$\epsilon(\mathbf{r}) = \bar{\epsilon} \mathbf{I} + \frac{2}{3} \epsilon_m^a \mathbf{Q}(\mathbf{r}). \quad (1)$$

Here,  $\bar{\epsilon}$  is the dielectric constant of the isotropic phase, and the molecular dielectric anisotropy  $\epsilon_m^a$  (for simplicity, taken to be constant) describes the effect of the nematic order on  $\epsilon$ . Specifying  $\epsilon$  in terms of  $\mathbf{Q}$  means that a specific nematic texture determines the profile of bound charges, which electrostatically couple to  $\rho_{\pm}(\mathbf{r})$ . Furthermore, it must be noted that specifying  $\epsilon = \epsilon(\mathbf{Q})$  describes –when inserted in the expression for the electrostatic free energy–, how the nematic director is affected by the presence of an electric field through the dielectric anisotropy. This so-called Fréederiksz effect [19] is important for the operation of most commercial liquid-crystal displays [20].

Besides the dielectric anisotropy, deforming a nematic texture can lead, depending on the properties of the mesogens, to a macroscopic polarization [21]. This so-called flexoelectric polarization  $\mathbf{P}_f$  is given in the one-constant approximation as

$$\mathbf{P}_f(\mathbf{r}) = G \nabla \cdot \mathbf{Q}(\mathbf{r}), \quad (2)$$

with  $G$  the flexoelectric constant. Strictly speaking, flexoelectric polarisation is associated with gradients in  $\hat{\mathbf{n}}(\mathbf{r})$  which leads to a non-zero  $\mathbf{P}_f(\mathbf{r})$ . Eq. (2) also contains the effects of order electricity, where gradients of the form  $\nabla S(\mathbf{r})$  result in a macroscopic polarisation. Here, we will use the term flexoelectric polarisation in a more loose sense, which includes the effects coming from an inhomogeneous texture in terms of both  $\hat{\mathbf{n}}(\mathbf{r})$  and  $S(\mathbf{r})$  (and biaxial order parameters). Furthermore, other gradients in  $\mathbf{Q}(\mathbf{r})$  should, in principle, be included in Eq. (2), such as  $Q_{\lambda\sigma}(\mathbf{r})\partial_{\alpha}Q_{\lambda\sigma}(\mathbf{r})$ ,  $Q_{\lambda\sigma}(\mathbf{r})\partial_{\sigma}Q_{\alpha\lambda}(\mathbf{r})$ , and  $Q_{\lambda\sigma}(\mathbf{r})\partial_{\alpha}Q_{\lambda\sigma}(\mathbf{r})$ , and  $Q_{\alpha\sigma}(\mathbf{r})\partial_{\lambda}Q_{\lambda\sigma}(\mathbf{r})$ , where we used the Einstein summation convention, which leads to the inclusion of flexoelectric modes that can vary in magnitude [22]. However, to reduce the number of free parameters, we choose the one-constant approximation, and in particular, the form of Eq. (2) because it is the simplest to implement numerically. Note that a flexoelectric polarisation produces a non-vanishing bound charge density,  $-\nabla \cdot \mathbf{P}_f(\mathbf{r})$  that is not produced via  $\epsilon(\mathbf{r})$ , to which ions couple through electrostatic interactions.

The final ion-nematic coupling that we introduce is an external potential for ions given by

$$V_{\pm}(\mathbf{r}) = k_B T g_{\pm} \text{Tr}[\mathbf{Q}(\mathbf{r})^2], \quad (3)$$

with  $g_{\pm}$  the dimensionless Gibbs transfer energies [23–25]. The above external potential quantifies the free energy cost of transferring an ion from an isotropic liquid to a nematic phase with given  $\mathbf{Q}(\mathbf{r})$ . In other words, the above contribution models the solvability difference between an ion in the nematic phase and an ion in the isotropic phase. We were the first to introduce such a contribution in the free energy for a nematic liquid crystal, see Ref. [A2]. The inspiration for including such an effect comes from models of electric double layers in binary fluid mixtures [26–29].

The Helmholtz free energy can now be written as

$$F[\rho_{\pm}, \psi, \mathbf{Q}] = F_{\text{N}}[\mathbf{Q}] + F_{\text{I}}[\rho_{\pm}] + F_{\text{E}}[\rho_{\pm}, \psi, \mathbf{Q}] + \sum_{i=\pm} \int_V d\mathbf{r} \rho_i(\mathbf{r}) V_i(\mathbf{r}), \quad (4)$$

with  $V$  the system volume. The nematic free energy is given by the standard Landau-de Gennes form

$$F_{\text{N}}[\mathbf{Q}] = \int_V d\mathbf{r} \left\{ \frac{L}{2} \partial_{\alpha} Q_{\beta\sigma}(\mathbf{r}) \partial_{\alpha} Q_{\beta\sigma}(\mathbf{r}) + \frac{A}{2} \text{Tr}[\mathbf{Q}(\mathbf{r})^2] + \frac{B}{3} \text{Tr}[\mathbf{Q}(\mathbf{r})^3] + \frac{C}{4} \{\text{Tr}[\mathbf{Q}(\mathbf{r})^2]\}^2 \right\}. \quad (5)$$

Here, nematic elasticity has been included within the one-constant approximation with elastic constant  $L$ . Furthermore,  $A$ ,  $B$ , and  $C$  are Landau-de Gennes parameters. Throughout our results, we use the standard values for these parameters of a typical liquid crystal (5CB) at room temperature. The ionic free energy  $F_{\text{I}}$  includes all non-electrostatic ion-ion and ion-mesogen effects, such as steric and other short-range interactions. However, for dilute ion concentrations, it suffices to use the ideal gas form (as in the well-known Poisson-Boltzmann theory) [18]

$$F_{\text{I}}[\rho_{\pm}] = k_{\text{B}}T \sum_{i=\pm} \int_V d\mathbf{r} \rho_i(\mathbf{r}) \{\ln[\rho_i(\mathbf{r})\Lambda_i^3] - 1\}. \quad (6)$$

Electrostatic interactions are encapsulated in the mean-field free energy

$$F_{\text{E}}[\rho_{\pm}, \psi, \mathbf{Q}] = \int_V d\mathbf{r} \left[ \mathbf{P}_{\text{f}}(\mathbf{r}) \cdot \nabla \psi(\mathbf{r}) + eq(\mathbf{r})\psi(\mathbf{r}) - \frac{1}{2} \epsilon_0 \epsilon_{ij}(\mathbf{Q}(\mathbf{r})) \partial_i \psi(\mathbf{r}) \partial_j \psi(\mathbf{r}) \right], \quad (7)$$

with  $\epsilon_0$  the vacuum permittivity and  $e$  the elementary charge unit. Note that we choose a form in which  $\psi(\mathbf{r})$  is a variational parameter [30]. In particular, taking  $\delta F_{\text{E}}/\delta \psi(\mathbf{r}) = 0$  results in the Poisson equation for an anisotropic dielectric with non-vanishing flexoelectric polarization. To compute the equilibrium profiles of  $\rho_{\pm}(\mathbf{r})$ ,  $\psi(\mathbf{r})$ , and  $\mathbf{Q}(\mathbf{r})$ , it is convenient to use the osmotic ensemble where the ions are treated grand canonically, i.e. the profiles are determined by

$$\min_{\rho_{\pm}, \psi, \mathbf{Q}} \left\{ F[\rho_{\pm}, \psi, \mathbf{Q}] - \sum_{i=\pm} \mu_i \int_V d\mathbf{r} \rho_i(\mathbf{r}) \right\}. \quad (8)$$

Here, the ionic chemical potentials are  $\mu_{\pm} = k_{\text{B}}T \ln(\rho_{\text{b}}\Lambda_{\pm}^3)$  with  $2\rho_{\text{b}}$  the *total* bulk ion reservoir density. In some cases, Eq. (8) can be computed analytically, but in most cases, the minimisation needs to be performed numerically. It must be noted that the separate free energy contributions in Eq. (4) are well studied. However, we are the first to combine these contributions to describe an ion-doped nematic fluid. We call the resulting theory Poisson-Boltzmann-Landau-de Gennes (PBLdG) theory, and we believe it is the simplest model to describe a nematic electrolyte. Results (i)-(iii) are associated with the equilibrium effects of this theory. Furthermore, Eq. (4) should be supplemented –depending on the geometry of interest – by some surface free energies, as we shall discuss in the description of the results, see Sec. C.3.

## II. Non-equilibrium effects

In result (iv), we investigated the effects of a nematic electrolyte out of equilibrium within the framework of linear irreversible thermodynamics [31]. In the case of nematic electrolytes, this framework incorporates the effects of ionic diffusion, the response to electrical fields, and fluid flow. In simple geometries, such effects have been experimentally studied in the context of, for example, non-linear liquid-crystal enabled electrophoresis [32, 33] and electroconvection rolls [34]. We contributed by incorporating the effects above in a self-contained theoretical framework, including the resulting out-of-equilibrium nemato-ionic couplings, which had not been studied in the literature before. Such

effects differ from equilibrium nemato-ionic couplings, like flexoelectricity or the dielectric anisotropy discussed in Sec. C.2.I, because they only occur out of equilibrium. To construct these non-equilibrium contributions, we use the principle of least dissipation [35],

$$\frac{\delta}{\delta \dot{q}_i} \left\{ \frac{D}{Dt} \left[ \frac{1}{2} \int d\mathbf{r} \rho_m \mathbf{v}(\mathbf{r}, t)^2 + F[\rho_{\pm}, \psi, \mathbf{Q}] \Big|_{\substack{\rho_{\pm}(\mathbf{r}) \rightarrow \rho_{\pm}(\mathbf{r}, t) \\ \psi(\mathbf{r}) \rightarrow \psi(\mathbf{r}, t)}}} \mathbf{Q}(\mathbf{r}) \rightarrow \mathbf{Q}(\mathbf{r}, t) \right] + \int_V d\mathbf{r} \mathcal{R} \right\} = 0. \quad (9)$$

Here  $\dot{q}_i$  is a short-hand notation for all generalised velocities of the system. Furthermore,  $D/Dt = \partial/\partial t + \mathbf{v} \cdot \nabla$  is the advective derivative with respect to the ‘‘barycentric’’ fluid velocity  $\mathbf{v}(\mathbf{r}, t)$ . The first term in square brackets in Eq. (9) is the kinetic energy for a fluid with mass density  $\rho_m$  which we assume to be incompressible,  $\nabla \cdot \mathbf{v} = 0$ . Finally,  $\mathcal{R}$  is the so-called Rayleighian, which incorporates dissipative out-of-equilibrium effects between the various fields.

In the literature, the above formalism was used to derive the equations for liquid-crystal-enabled electrophoresis in terms of  $\mathbf{Q}$  [36]. However, in this work, the dissipative nemato-ionic couplings were neglected. To have a simplified picture of such phenomena, we investigated these effects within the uniaxial approximation where  $\mathbf{Q} = (3/2)S[\hat{\mathbf{n}}\hat{\mathbf{n}} - (1/3)\mathbf{I}]$ , with  $S$  spatially constant. Furthermore, in Ref. [A5], we focused on the situation where  $\mathbf{v} = 0$  to single out the new electrokinetic effects independent from the fluid flow. In this picture, we can construct the total Rayleighian as

$$\mathcal{R} = \mathcal{R}_N + \mathcal{R}_I + \mathcal{R}_C. \quad (10)$$

Here, the nematic Rayleighian is given by

$$\mathcal{R}_N = \frac{\alpha}{2} [\partial_t \hat{\mathbf{n}}(\mathbf{r}, t)]^2, \quad (11)$$

with  $\alpha$  the rotational viscosity. For the ionic part, we take the diffusive form

$$\mathcal{R}_I = \frac{1}{2} k_B T \sum_{i=\pm} \rho_i(\mathbf{r}, t) \mathbf{u}_i(\mathbf{r}, t) \cdot \mathbf{D}_i^{-1} \cdot \mathbf{u}_i(\mathbf{r}, t), \quad (12)$$

with  $\mathbf{D}_{\pm}$  the (anisotropic) ionic diffusion tensor, neglecting cross-diffusion effects. The dissipative coupling of ionic fluxes with the nematic background is captured by

$$\mathcal{R}_C = \sum_{i=\pm} \gamma_i \rho_i(\mathbf{r}, t) \partial_t \hat{\mathbf{n}}(\mathbf{r}, t) \cdot \{[\mathbf{u}_i(\mathbf{r}, t) \cdot \nabla] \hat{\mathbf{n}}(\mathbf{r}, t)\}, \quad (13)$$

with phenomenological ‘‘friction’’ coefficients  $\gamma_{\pm}$ . Note that in writing  $\mathcal{R}_C$ , we only include terms which are invariant under independent rotations of real space and the nematic director. Such an artificial symmetry is akin to the equal-constant approximation that is frequently used for the Frank elastic free energy. In principle, more terms can be added, but we focus here on the simplest contributions.

For the given  $\mathcal{R}$  the generalised velocities are  $\mathbf{u}_{\pm}$  and  $\partial_t \hat{\mathbf{n}}$ . Minimisation with respect to  $\mathbf{u}_{\pm}$  by using Eq. (9) gives

$$\mathbf{u}_{\pm}(\mathbf{r}, t) = -\frac{1}{k_B T} \mathbf{D}_{\pm} \cdot \left[ \nabla \frac{\delta F}{\delta \rho_{\pm}(\mathbf{r}, t)} + \mathcal{E}_{\pm}(\mathbf{r}, t) \right], \quad (14)$$

where an additional ‘‘motive’’ force is generated given by

$$\mathcal{E}_{\pm}(\mathbf{r}, t) = \gamma_{\pm} \nabla \hat{\mathbf{n}}(\mathbf{r}, t) \cdot \partial_t \hat{\mathbf{n}}(\mathbf{r}, t). \quad (15)$$

Eq. (14) should be combined with the continuity equation  $\partial_t \rho_{\pm}(\mathbf{r}, t) + \nabla \cdot [\rho_{\pm}(\mathbf{r}, t) \mathbf{u}_{\pm}(\mathbf{r}, t)] = 0$ . Note that for isotropic ionic diffusion and  $\mathcal{E}_{\pm} = 0$  that this result is just the well-known Poisson-Nernst-Planck theory for the electrokinetics of an isotropic electrolyte [37]. Variation with respect to  $\partial_t \hat{\mathbf{n}}$  gives

$$\alpha \partial_t \hat{\mathbf{n}}(\mathbf{r}, t) = -[\mathbf{h}_{\perp}(\mathbf{r}, t) + \mathbf{h}_d(\mathbf{r}, t)]. \quad (16)$$

Here, the molecular field is given by  $\mathbf{h}_\perp = \hat{\mathbf{n}} \times \delta F / \delta \hat{\mathbf{n}} \times \hat{\mathbf{n}}$ , where the vector products are added to ensure the normalisation  $|\hat{\mathbf{n}}| = 1$ . The dissipative field is given by

$$\mathbf{h}_d(\mathbf{r}, t) = \sum_{i=\pm} \gamma_i \rho_i(\mathbf{r}, t) \{[\mathbf{u}_i(\mathbf{r}, t) \cdot \nabla] \hat{\mathbf{n}}(\mathbf{r}, t)\}. \quad (17)$$

Note that the generalized forces  $\mathcal{E}_\pm$  and  $\mathbf{h}_d$  are Onsager reciprocal with generalized fluxes  $\partial_t \hat{\mathbf{n}}$  and  $\rho_\pm \mathbf{u}_\pm$ . They are the new couplings that we introduced in Ref. [A1]. In Sec. C.3.IV, we discuss some of the repercussions of these reciprocal nemato-ionic couplings.

### 3. Description of results

#### I. Electric field effects on anchoring

In article [A4], we investigated the effects of a charged surface and its accompanying double layer on the surface anchoring. Anchoring is the surface-imposed ordering of particles close to an external surface, such as a polymer-rubbed wall or glass [38]. Using different types of materials with a variety of physicochemical treatment of the surfaces, one can realise surface orientational ordering in almost every arbitrary direction, like planar [39], perpendicular (homeotropic) [40], tilted orientations [41], including surface patterning of a combination of these (see also Sec. C.3.III). In technological applications, manipulating surface anchoring is valuable in producing displays [20], sensors [42], and microfluidics [43]. Methodologically speaking, the effects of surface anchoring can be included in the formalism of Sec. C.2. by including an appropriate surface free energy term, which on the level of Euler-Lagrange equations imposes boundary conditions on the order parameter. A popular surface free energy is the so-called Rapini-Papoular form [44],

$$F_S[\mathbf{Q}] = \frac{W}{2} \int_{\Omega} dS \operatorname{tr}[(\mathbf{Q} - \mathbf{Q}^0)^2], \quad (18)$$

for given anchoring strength  $W$  and imposed surface order parameter  $\mathbf{Q}^0$ . The integral is over a surface  $\Omega$ . Combined with other terms in the free energy, the surface order parameter is determined by the combination of nematic elasticity, given by the value of the elastic constant(s), the value of  $W$ , and the system size. When this surface is electrically charged, there will be additional effects due to the electric field produced by the charged surface.

Here, we focused on a slab geometry confined by two parallel flat plates, which can be modelled in one spatial dimension due to the in-plane translational invariance. We consider the situation where the bottom surface is charged and described by the Rapini-Papoular surface free energy with planar orientation (finite value of  $W$ ), whereas the top surface is uncharged with strong homeotropic anchoring (formally understood as  $W \rightarrow \infty$ ). Surface anchoring biases the director to be parallel to the bottom plate, which induces an elastic distortion in the cell since  $\hat{\mathbf{n}}$  is always perpendicular to the top surface. In contrast, nematic elasticity directs the system towards a state where the elastic distortion is minimised and, thus, where the director is perpendicular to the bottom plate. In other words, the effects of anchoring and elasticity compete in this geometry.

The charged bottom surface produces an electric field perpendicular to both plates. When we include only the dielectric anisotropy, this gives an electrostatic free energy density of the form  $\sim -\Delta\epsilon(\hat{\mathbf{n}} \cdot \mathbf{E})^2$  with  $\mathbf{E}$  the local electric field, and  $\Delta\epsilon = \epsilon_m^a S$  (see Eq. (1)) the dielectric anisotropy coefficient. When  $\Delta\epsilon > 0$ , the corresponding dielectric torque pushes the local director to be perpendicular to both surfaces. In the presence of ions, this electric field is screened, and one can tune the range of the electric field by the ion content, as set by the Debye screening length.

We have systematically investigated the interplay of a (screened) electric field, nematic elasticity, and surface anchoring in article [A4] by determining the director's orientation at the bottom plate

in this specific geometry. The relevant Euler-Lagrange equations were numerically solved using finite-element methods. Furthermore, we have also considered the effects of so-called charge regulation [45], where the surface charge density on the bottom plate is a function of the ion content and, thus, the Debye length of the system. Such a boundary condition is microscopically realised by chargeable chemical groups on the surface of interest. Interesting phenomena occur when increasing the Debye length enlarges the range of the electric field but reduces the surface charge, which gives non-monotonous effects of the bottom surface director orientation as a function of the Debye length. Finally, we have also explored the effects of flexoelectricity, which produces an additional polarisation field that affects the local director profile. Taken together, we conclude that it is possible to tune the surface ordering by the salt content in the system –provided it can be carefully experimentally controlled (which can be a challenge in practical situations).

## II. Anisotropic screening

In electrostatics, it is well known that a charged particle set in the origin of our coordinate system produces an electrostatic potential that asymptotically decays like  $\psi(\mathbf{r}) \sim 1/r$  for  $r \rightarrow \infty$ , irrespective of the particle shape. When such a particle is accompanied by an electric double layer, for example, by immersing the particle in water, the situation changes, and the asymptotic relation is altered to [46–48]

$$\psi(\mathbf{r}) \sim \mathcal{A}(\varphi, \theta) \frac{\exp(-r/\ell_D)}{r}, \quad (r \rightarrow \infty), \quad (19)$$

with  $\ell_D$  the Debye length. Here,  $\mathcal{A}(\varphi, \theta)$  is the so-called anisotropy function, which depends on the azimuthal angle  $\varphi$  and polar angle  $\theta$ . Furthermore,  $\mathcal{A}(\varphi, \theta) = 1$  for a sphere, but for any other non-spherical particle shape, it inherits the “shape” of the particle, which is important even infinitely far from the particle, in contrast to the unscreened case. Note that there is only a single decay length  $\ell_D$  which does not depend on any angle.

In Article [A3], we investigated how Eq. (19) is altered when the particle is isotropic (i.e., a sphere), but the medium is anisotropic: in our case, a nematic fluid with a spatially constant director field. Using Poisson-Boltzmann theory with an anisotropic dielectric tensor parametrised in the director picture as  $\boldsymbol{\epsilon} = \epsilon_{\perp} \mathbf{I} + \Delta\epsilon \hat{\mathbf{n}}\hat{\mathbf{n}}$ , with  $\Delta\epsilon = \epsilon_{\parallel} - \epsilon_{\perp}$ , we have computed the asymptotic behaviour of  $\psi(\mathbf{r})$ . We find the explicit relation

$$\psi(\mathbf{r}) \sim \tilde{\mathcal{A}}(\theta) \frac{\exp[-r/\ell_D^N(\theta)]}{r}, \quad (r \rightarrow \infty). \quad (20)$$

Similar to an anisotropic particle in an isotropic medium, the asymptotic decay of  $\psi(\mathbf{r})$  is modulated by an anisotropy function  $\tilde{\mathcal{A}}(\theta)$ , which from symmetry reasons only depends on the angle  $\theta$  between the position vector  $\mathbf{r}$  and the spatially uniform director  $\hat{\mathbf{n}}$ . Different from Eq. (19), however, is the appearance of an anisotropic Debye length, given by the exact expression

$$\ell_D^N(\theta) = \ell_D \sqrt{\frac{\epsilon_{\perp} \epsilon_{\parallel}}{\bar{\epsilon}(\epsilon_{\parallel} - \Delta\epsilon \cos^2 \theta)}}. \quad (21)$$

Here,  $\ell_D$  should be interpreted as the Debye length of a “hypothetical” isotropic system at the same bulk ion concentration and temperature. When we compare the electric double layer surrounding a charged sphere in a nematic medium with  $\epsilon_{\parallel} > \epsilon_{\perp}$  with that of a prolate spheroid in an isotropic medium, we notice that the isopotential lines close to the particle surface have a similar shape in both cases. However, the asymptotic decay of  $\psi(\mathbf{r})$  is vastly different, compare Eq. (19) with Eq. (20), due to the appearance of anisotropic Debye length given by Eq. (21). In Ref. [A3], we also obtained accurate analytical expressions for  $\psi(\mathbf{r})$  valid for all  $\mathbf{r}$  by using a semi-analytical method that we have developed [49] combined with a multipole expansion.



Unfortunately, within the current state-of-the-art, no direct measurements are possible for an electric double layer of a single particle. Therefore, to expand upon the experimental relevance, we computed the effective interaction potential between a pair of equal-sized charged spheres to see the effects of an anisotropic double layer. Here, we used again the theory from Ref. [49] combined with a multipole expansion. Furthermore, by comparison with numerical finite-element calculations, we have shown that this method yields accurate expressions for the effective pair potential, with deviations from the numerical results only occurring when the particles are close to contact. In practice, it is difficult to experimentally realise a uniform director field around spherical particles. Therefore, we tested our theory with experiments for the case where a Saturn-ring defect surrounds the particles. In the uncharged case, such a defect causes quadrupolar effective elastic pair interactions, for which analytical expressions also exist within a multipole expansion [40]. Therefore, within the assumption of linear additivity, one can add the elastic interaction and the van der Waals interaction to our calculated screened Coulomb effective interactions with anisotropic double layers. Furthermore, we used an approximation that did not include elastic distortions in the effective electrostatic interactions.

The resulting theory predicts a shallow minimum for sufficiently large interparticle separations on the order of one to several particle diameters. Such widely separated metastable particle states do not exist for a pair of charged particles in an isotropic liquid –as is well known from the celebrated Derjaguin-Landau-Verwey-Overbeek (DLVO) theory [50, 51]– due to the absence of the (attractive) nematic elastic interactions. Furthermore, our theory showed that the metastable equilibrium angle of the separation vector with the (far-field) uniform director field is shifted by a few degrees compared to the case of isotropic screening. Although seemingly a small effect, we experimentally resolved the metastable equilibrium configuration in article [A3], where the angular deviation from a hypothetical case of isotropic electrostatic screening in a nematic fluid was approximately five degrees.

### III. Structure of electric double layers in nematic fluids

As is common with symmetry-broken phases of matter, nematic fluids can host regions of topologically protected patterns called topological defects [52], where the underlying order parameter is frustrated. In the case of bulk nematic fluids, the presence of a topological defect implies that there is a region where the elastic distortions are locally singular. To escape from such a high elastic free energy cost, the nematic order in the core of a defect melts into an isotropic core, where the scalar order parameter  $S$  is locally reduced from the nematic bulk value [53]. In contrast to ferromagnets, nematic defects can carry half-integer values of the topological invariants due to the residual  $\mathbb{Z}_2$  symmetry [1]. Furthermore, in liquid crystals, the current experimental techniques allow for a high degree of spatial manipulation of defects, with the use of electric [54], magnetic [55], and optical fields [56], but also fluid flow [57]. Combined with the experimental tools of realising various topological defects in nematic fluids, such as loops, points, solitons, and even knots [58–60], it is clear that topological defects form an indispensable tool for manipulating the anisotropic properties of a liquid crystal.

In Article [A2], we investigated whether topological defects in a bulk nematic fluid could function as regions of local ionic charge separation. In bulk one-component isotropic liquids, this cannot occur because any local charge separation is realised through the formation of electric double layers, which needs the presence of charged surfaces, such as charged walls or charged colloidal particles [61]. In liquid crystals, however, there are topological defects. The presence of a molten isotropic core and the large elastic distortions surrounding a defect, naturally introduce nemato-ionic couplings via flexoelectricity (Eq. (2)) and ion solvability differences (Eq. (3)), respectively.

Using a combination of analytical and numerical methods of the full PBLdG theory discussed in Sec. C.2.I, we investigated a few geometries where ionic charge separation can occur. The simplest (one-dimensional) geometry is that of an isotropic-nematic interface. We derived analytical expressions for the ion density and potential profiles in this case. We found that a back-to-back electric double

layer forms across the isotropic-nematic interface similarly to an ionically charged oil-water interface [62]. Differently here is that also order electricity can play a role, which can shift the the sign and magnitude of charges formed at both sides of the interface.

For all other geometries, we resorted to numerical finite-element calculations. Specifically, we explored ionic charge separation in radial hedgehog defects [63], half-integer wedge disclinations [64], and finally, the Saturn-ring defect and point defect that forms around a spherical colloidal particle with homeotropic anchoring [40]. In all cases, we found that the ionic charge density profile follows the profile of bound charges produced by flexoelectricity when the Debye length is sufficiently small. However, the charge profiles are broadened when the Debye length is larger. In other words, for a small number of ions, the entropic cost of following the flexoelectric pattern is too small compared to the benefit of satisfying local charge neutrality. However, this is not a problem when a sufficiently large number of ions are present. On the other hand, the coupling introduced in Eq. (3) charges the isotropic core of a defect (for  $g_{\pm} > 0$ ) because ions are generally speaking more soluble in the isotropic phase. The resulting ion density profiles result from these two effects, i.e., from flexoelectricity and ion solvability differences. With these effects, we found that, in contrast to an isotropic electrolyte where only electric double layers are formed, that a (bulk) nematic electrolyte can even host electric multilayers where three or more regions of positive and negative charges meet.

In article [A2], we focused on diffuse layers of ions in a nematic texture. However, as we know from the behaviour of ions in isotropic liquids, an electric double layer can also involve the presence of charged surfaces. In article [A5], we showed that topological defects can influence the local surface charge distribution of external surfaces, like a charged wall or a charged colloidal particle. The simplest geometry for demonstrating this phenomenon is a slab geometry consisting of two parallel walls. Different from the relatively simple director configurations in article [A4], we now used a charged bottom plate where we fixed a two-dimensional nematic surface pattern via strong anchoring conditions. The top plate is uncharged with tunable homeotropic anchoring strength  $W$ . The nematic pattern on the bottom plate is chosen such that for small values of  $W$ , there is a surface defect on the top plate, whereas, for larger values of  $W$ , there is a bulk  $-1/2$  defect within the slab with a tunable position depending on the magnitude of  $W$ . This geometry models the situation as if we could quasi-statically move the defect from the surface of the top plate towards the bottom plate using external fields. When we impose charge regulation boundary conditions [45] on the bottom plate, such as constant-potential boundary conditions or taking some ion adsorption isotherm of the Langmuir form, we find that moving the defect closer to the bottom surface locally increases or decreases the surface charge distribution depending on the proximity to the topological defect. This effect occurs even when there is just the dielectric anisotropy, but is more pronounced when flexoelectricity is included.

Furthermore, we investigated the generality of this physical phenomenon in different geometries. We found that it is possible to manipulate the surface charge distribution on an external wall or particle when there is a perpendicular director gradient that varies across the surface. With this principle, we showed in article [A2] that even two-dimensional inhomogeneous surface charge distributions can be created in a slab geometry using defect lines that connect both plates. Furthermore, we showed that the concept persists in charged colloidal particles, by demonstrating that a Saturn-ring defect and a point defect cause significantly different surface charge distributions around a charged colloidal sphere.

#### IV. Reciprocal effects of electrokinetics with nematic texture dynamics

In article [A1], we investigated what physical effects are present when a nematic electrolyte is driven out of equilibrium with charge currents. To obtain analytical insights, we took the dynamical equations derived in Sec. C.2.II and focused only on the so-called charge sector  $e[\rho_+(\mathbf{r}) - \rho_-(\mathbf{r})]$ . We neglected the osmotic sector  $\rho_+(\mathbf{r}) + \rho_-(\mathbf{r})$  by expanding the equations around a uniform bulk density and keeping only the first-order contribution. In this simplified picture, the complete dynamics are

determined by the generalised fluxes  $\partial_t \hat{\mathbf{n}}$  and the ionic charge current  $\mathbf{j}$ . In addition to the driving forces that come from the free energy of the system, we have the dissipative driving forces

$$\mathbf{h}_d = \gamma(\mathbf{j} \cdot \nabla) \hat{\mathbf{n}}, \quad \mathcal{E} = \gamma(\nabla \hat{\mathbf{n}}) \cdot (\partial_t \hat{\mathbf{n}}). \quad (22)$$

Here, there is only one dissipative coupling constant  $\gamma$  compared to the two coupling constants  $\gamma_{\pm}$  when the osmotic sector is included, see Sec. C.2.II.

In general, the dissipative field  $\mathbf{h}_d$  introduces a ‘‘torque’’ on the nematic director when a charge current  $\mathbf{j}$  is passed through a nematic texture. Consequently, an ionic current in a nematic fluid can drive a nematic texture. Alternatively, movement of nematic texture leads to the formation of a charge current because ions are pumped by the motive force  $\mathcal{E}$ . These two reciprocal effects are similar to the concepts of spin torque and spin pumping, respectively, within the field of spintronics [65]. This scientific field focuses on manipulating the quantum-mechanical electron spin by exploiting electronic charges for device applications. Here, we have produced the nematic analogue of the spin torque and spin pumping, where the spin is replaced by the nematic director and the electronic charges by ionic charges. The resulting dynamics, which emerge from the reciprocal nemato-ionic dissipative couplings, are similar to how an Archimedes’ screw functions: a hydraulic machine that either pumps water or generates energy as a turbine [66].

We focused first on a slab geometry of two parallel plates to show the consequences of Archimedes’ screw-like dynamics. Here, we imposed strong anchoring conditions on both plates, with planar anchoring on the bottom plate and homeotropic anchoring on the top plate. In the quasistatic situation, where we pass a slowly varying current through the sample, it turns out that the nematic texture couples to  $\mathbf{j}$  through the winding density  $\eta(z) = (\hat{\mathbf{n}} \times \partial_z \hat{\mathbf{n}}) \cdot \hat{\mathbf{x}}$ . In this expression, we assumed that  $\hat{\mathbf{n}}$  lies in the  $yz$  plane. From the explicit quasistatic solution of the underlying equations, we find that the winding density is dragged along the direction of  $\mathbf{j}$ . This effect can be qualitatively distinguished from the Fréedericksz effects stemming from the  $-\Delta\epsilon(\hat{\mathbf{n}} \cdot \mathbf{E})^2$  term in the free energy which does not depend on the sign of  $\mathbf{j}$ . From the backreaction of the nematic dynamics on the electrical response, we find that the motive force  $\mathcal{E}$  causes an effective electric potential difference between the two plates that is of the inductive form. The resulting impedance is experimentally accessible and shows that topological defects in a nematic cell between two electrodes can carry inductance for non-zero  $\gamma$ .

As a second way to measure  $\gamma$ , we proposed in article [A1] a setup where the core of a  $+1/2$  disclination can be moved with a charge current. This situation differs from the known examples in the literature, where defect movement is induced via fluid flow [67]. In this specific geometry, we found an analytical expression for the terminal velocity of a disclination, and we have shown that it depends linearly on  $\mathbf{j}$  with proportionality constant  $\gamma$ . Thus studying the defect movement for varying  $\mathbf{j}$  allows us to experimentally measure the unknown value of  $\gamma$ .

#### 4. Summary

In conclusion, we have shown some examples where electric double layers in anisotropic nematic fluids play an important role in equilibrium and out-of-equilibrium situations. In particular, we have highlighted how a nematic texture dictates the spatial profiles of ionic charges around various director configurations, like topological defects, and how surface charge distributions of external walls and particles can be manipulated via the nematic texture in contact with these objects. Furthermore, we have shown how electrostatics can influence the surface anchoring of mesogens and how the dielectric anisotropy of a nematic medium affects electrostatic screening. For out-of-equilibrium situations, we have highlighted the reciprocal nature of nemato-ionic dissipative couplings, which lead to a nematic torque induced by charge currents, or reciprocally, ion pumping via nematic texture movement.

Because of the pioneering nature of our studies, there are many possibilities for future research. I received a Sonata Bis grant from the Polish National Science Centre to pursue new ideas in this

direction. In particular, I am interested in the effects of higher ion concentrations and electric fields beyond the Poisson-Boltzmann approximation, where ion-ion and ion-mesogen steric effects play an important role. In this context, I am interested specifically in utilizing nematic electrolytes in energy-storage applications, such as nematic-based electric double-layer capacitors. Furthermore, I am excited to explore the effects of ions in the recently discovered nematic ferroelectrics [68], which might have promising electro-optical applications due to their large macroscopic polarisation combined with a fluid-like nature. Finally, on the level of out-of-equilibrium situations, I am interested in expanding upon the reciprocal nemato-ionic couplings we introduced in our work, and studying their interplay with fluid flow and activity.

In a broader scope, I plan to broaden my research portfolio towards other topics as well. I have started taking steps in this direction by working on chiral active fluids and Brownian motion in many-body systems. As a far-reaching goal, I am very much interested in how the ionic phenomena in this achievement could be used for so-called neuromorphic computing, which involves brain-inspired computational methods. Here, it is believed that ions in soft materials can play an important role in the development of this field [69, 70]. Furthermore, I plan to move towards biological physics, where I think there is much unexplored territory at the moment. Currently, I am working on a project to quantify the effects of nematically ordered charged DNA in virus capsids on effective interactions. Finally, I envision many possibilities when soft-matter systems are combined with quantum systems [71–73], which could lead to exciting new phenomena.

## **5 PRESENTATION OF SIGNIFICANT SCIENTIFIC OR ARTISTIC ACTIVITY CARRIED OUT AT MORE THAN ONE UNIVERSITY, SCIENTIFIC OR CULTURAL INSTITUTION, ESPECIALLY AT FOREIGN INSTITUTIONS**

Already at an early stage in my career, I was involved in scientific research. My first research project started during my B.Sc. studies when I was hired as a junior researcher at the Spinoza Institute at the Eindhoven University of Technology. This institute was founded by prof. Bert Meijer but led by prof. Anja Palmans. During this period, I worked on two projects related to supramolecular self-assembly, which led to two publications to which I made a significant contribution. The main objects of study were disc-like molecules (so-called benzenetricarboxamides, or BTAs) that were known to self-assemble into long helical fibres in dilute solution. Under normal circumstances, i.e., when the molecular building blocks are achiral, both left and right-handed helices form in equal amounts. However, when the disc-like molecules are chiral –meaning that the mirror image of the three-dimensional molecular structure cannot be superimposed on itself– the chirality on the molecular level is “transferred” to the mesoscopic level of a helix, and only fibres with one type of handedness are formed. My main tasks (under the daily supervision of Maarten Smulders and Anja Palmans) were to synthesize and characterize a specific type of BTA with a different amide connectivity than other known BTAs in the literature. Subsequently, I analyzed this molecule in dilute solution when mixed with other types of (a)chiral BTAs and characterized the amount and kind of helical fibres formed in dilute solution using spectroscopic methods. Later, Patrick Stals also analysed the crystal structure of such BTAs, and Ivo Filot analysed the structure of helices using quantum-chemical methods. Later, this joint work led to a publication in *Chemistry: A European Journal*.

For my B.Sc. thesis, I experimentally analysed a different type of BTA in a similar way (together with Seda Cantekin and Yoko Nakano), where the chirality was introduced via isotope substitution of hydrogen atoms by deuterium. For such a molecule that is chiral in a “minimal” way, we found that the solvent plays an important role in the self-assembly properties, which is uncommon for such systems. For example, the solvent type can alter not only the handedness of the helical fibre but also

its structural properties. Furthermore, I worked with prof. Paul van der Schoot on the theory of self-assembly for such structures. Ultimately, this work was published in Chemical Communications.

After these experimental projects that were mainly focused on chemistry, I moved to theoretical physics. During my M.Sc. thesis project, I worked under the supervision of prof. Cristiane de Moraes Smith at Utrecht University on realising topological insulators in two-dimensional lattices. Within a tight-binding description, the kinetic energy of fermions or bosons occupying the lattice sites is described by so-called hopping terms in the (effective) Hamiltonian. Such contributions emerge from the overlap of chosen wave functions that are localised on the lattice sites. Under normal circumstances, the nearest-neighbour hopping parameter  $t$  is larger than the next-nearest-neighbour hopping parameter  $t'$ . However, in experiments with cold atoms in periodically time-modulated optical lattices, one can effectively obtain the situation where  $t' > t$ . Using  $t'$  as an adjustable parameter, we found together with Wouter Beugeling that it is possible to change the Chern number of a specific band, provided that the unit cell of the lattice contains more than two lattice sites. This Chern number is directly related to the number of conducting states on the boundary of a finite lattice –the so-called edge states–, given that the bulk is an insulator. The observation that tuning  $t'$  can change the number of edge states of a bulk insulating system led to a publication in Physical Review B.

For my Ph.D. studies, I changed my subject from hard condensed matter to soft condensed matter. During this period, I worked under the supervision of prof. René van Roij at Utrecht University on the physics of charged colloidal particles. Such particles (with sizes on the order of a few nanometers to microns) are large enough that they can be experimentally studied using optical microscopes but small enough that they exert Brownian motion. When these particles are brought into contact with a solvent containing ions, an electric double layer forms. If water is the solvent, the Debye length is generally much smaller than the size of a colloidal particle. At the time, the group of prof. Alfons van Blaaderen (with whom we collaborated) could also realise charged colloidal particles in oily solvents. Oily solvents are less polar than water, and it is, therefore, much more challenging to dissolve ions in them. However, a small number of ions can still be dissolved in some particular oils. Consequently, electric double layers can be realised with thicknesses on the order of, or even larger than, the typical size of a colloidal particle. Electrostatic interactions in such colloidal suspensions play a more important role than in charged colloids based on water because of the weaker electrostatic screening.

Inspired by this experimental work, we used Poisson-Boltzmann-type methods to compute the equation of the state of charged colloids where the Debye length and colloid density are taken as tunable parameters. In collaboration with Niels Boon, we utilized a suitable mapping of the suspension within the spherical cell approximation to a one-component plasma (which is adequate for charged colloids with large double layers) to obtain reliable results. With this methodology, we could explain the reentrant melting of charged colloidal particles, where a sequence of fluid  $\rightarrow$  body-centred cubic crystal  $\rightarrow$  fluid was found upon increasing the colloid density at fixed ion concentration. We found that the essential ingredient for such behaviour was that the surface chemistry of the particles allowed for the adsorption of positive and negative ions. This should be contrasted with the single-ion adsorption case, where no reentrant melting is present. Our findings were published in Physical Chemistry Chemical Physics. Furthermore, using again Poisson-Boltzmann type methods, we were able to explain the formation of alternating strings and clusters in mixtures of equally-charged colloidal particles (experimentally observed by Marjolein van der Linden from the van Blaaderen group) using a careful analysis of the effective interaction potential between two charged colloidal particles. Here, again, the surface chemistry matters, which enabled the formation of patches with the opposite charge on the particle surface upon decreasing the average inter-particle distance, which led to an effective anisotropic attraction. We reported our results in Soft Matter.

The second part of my Ph.D. work focused on the behaviour of oil-dispersed charged colloidal particles near oil-water interfaces. Often, it is stated in the literature that such particles –when

adsorbed to an oil-water interface— cannot be detached because of the wetting interaction energies that are on the order of  $\sim 10^6 k_B T$ . However, experiments performed by Nina Elbers and Jessi van der Hoeven in the group of prof. Alfons van Blaaderen showed that systems can be realised where the particles are close to the oil-water interface without actually intersecting the interface. A competition between a repulsive particle-oil-water van der Waals interaction with screened Coulomb interactions stabilised such a configuration. In this case, the particles can be detached from the oil-water interface by adding ions to the oil phase. Using a modified Poisson-Boltzmann theory, we theoretically modelled such scenarios with Sela Samin by numerically computing the effective interaction potential between a charged spherical particle and a flat oil-water interface. We have shown how this interaction potential depends on the surface chemistry of the particle and the charge of the oil-water interface. With this theoretical model, we were able to explain the experimental observations. The ion-induced detachment from the interface could then be rationalised from the charge reversal of the colloidal particle from positively charged to negatively charged, which then interacts with a charged oil-water interface that is not charge reversed. This effect can become stronger than the image-charge interaction between particle and interface that stems from the dielectric contrast between the particle, oil, and water phases, which in the typical experimental scenario of interest always leads to attraction. This work led to a theoretical paper in *Physical Review Letters* and a paper with experimentalists in *Physical Chemistry Chemical Physics* (the latter published after obtaining my Ph.D.).

Apart from my work on charged colloids, I also contributed to the field of liquid crystals during my Ph.D. studies by developing a Landau-de Gennes theory for hard colloidal rods. This work has been performed with Melle Punter, Sela Samin, Paul van der Schoot, and René van Roij. “Usual” Landau-de Gennes theories are typically geared towards so-called thermotropic liquid crystals, where nematic order emerges upon lowering the temperature. Hard colloidal rods, however, form liquid crystalline phases upon increasing the particle density, and the phase transition is accompanied by a density jump. We proposed an accurate way to account for such a density jump by using the chemical potential as a control parameter instead of the temperature. Furthermore, we have shown how this methodology performs under specific situations, like the size of a topological defect, liquid crystals under confinement, and the formation of liquid-crystalline droplets (the so-called tactoids).

During my Ph.D. studies, I focused mainly on equilibrium scenarios involving electric double layers. However, in later years, I also worked with prof. René van Roij on non-equilibrium situations. Specifically, I contributed by extending the effects of surface chemistry to out-of-equilibrium situations using electrokinetic equations with suitable boundary conditions. Using this methodology, Ben Werkhoven has then shown that fluid flow can affect the surface charge density distribution of an ion-adsorbing wall. These findings were published in *Physical Review Letters*. Non-equilibrium effects were also crucial in the sedimentation of a binary mixture of equally charged colloidal particles. Here, using a simple effective dynamics, we found quantitative agreement with experiment for the stationary density profiles of the various types of colloidal particles. Furthermore, we were able to explain the occurrence of a so-called Brazil-nut effect, where heavier particles “float” on top of the lighter particles. The equilibrium situation is determined by the charge ratio of the two types of particles compared to the ratio of their buoyant masses. We have shown using theoretical methods that the experimentally observed overall density dependence of this phenomenon (experiments performed by Marjolein van der Linden) turns out to be a question of whether the equilibrium situation is reached. We published the explanation and observation of the colloidal Brazil-nut effect in the *Proceedings of the National Academy of Sciences U.S.A.*

During my first postdoc at the University of Ljubljana, Slovenia, I initially continued working on charged colloids. In particular, I have worked with prof. Miha Ravnik on the structure of electric double layers within Poisson-Boltzmann theory for more complicated particle shapes than spheres. In particular, we were interested in the topological shapes of surfaces of constant charge density when

the particle has a non-trivial topology. We analysed the topological properties of such isosurfaces as a function of the Debye length. This work was published in Scientific Reports. Later, the effect of the shape of the electric double layers on the effective pair potential between two particles was studied. From this idea, I have developed an efficient approximate method for analytically computing the interaction potential between two particles for arbitrary particle separations and orientations by utilizing a suitable mapping of the particle to a singular charge distribution. This work led to a single-author publication in Physical Review Research and formed the basis for the analytical methods employed in Ref. [A3].

In my second postdoc, with prof. Robert Hołyst at the Institute of Physical Chemistry, Warsaw, Polish Academy of Sciences, I worked on the self-diffusion of a spherical probe particle in a complex liquid composed of crowder particles. We are currently preparing manuscripts for our findings with Karol Makuch. In particular, we were interested in obtaining an analytical expression for the diffusion constant as a function of the probe size for a given interaction potential between the probe and crowd-ers. This is a very complex problem since one needs to bridge orders of magnitudes in length scales using theoretical methods. Our most important result is that we have shown that the ratio of the long-time self-diffusion constant to the single-particle diffusion constant,  $D/D_0$ , does not approach unity for the point probe limit. However, this is often assumed in the literature when constructing empirical formulas. Here, the point limit should be understood as the situation where the probe size is much smaller than the crowder size but still bigger than a solvent particle. Instead,  $D/D_0$  goes to a value that depends on the type of crowder-probe interaction potential, which can be explicitly computed. This physical limit forms a firm basis for constructing empirical expressions to describe experimental data for all probe sizes. The manuscripts are currently in preparation because we are waiting for some new experimental results and data analyses, which should be modified with these latest insights. However, I feel it is adequate to summarize such findings here despite not having published on this subject yet, considering the amount of time I spent at the Institute of Physical Chemistry as a postdoc.

Finally, when I moved to the University of Warsaw, I worked with prof. Bogdan Cichocki on incompressible three-dimensional chiral active fluids. Such fluids, like liquid crystals, are anisotropic in nature. However, the main difference with liquid crystals is that the anisotropy stems from self-spinning particles subjected to active torques. Consequently, the order parameter –the intrinsic angular momentum density – is odd under time reversal, contrasting e.g. the nematic director, which is invariant under time reversal. Therefore, the viscosity tensor of such a fluid has a richer structure than “ordinary” liquids because of a non-vanishing antisymmetric part, also known as odd viscosity. For a simplified model with only one shear and one odd viscosity, we have explicitly computed the exact Green’s function for such fluid in the creeping flow regime. Furthermore, we solved the single-particle problem for a sphere undergoing solid-body motion with stick boundary conditions in linear ambient flow. In particular, we have analytically computed the full grand mobility matrix for such a system and analysed the consequences, such as the effective viscosity tensor of a suspension of passive particles immersed in an odd viscous fluid. This work has been recently published in Physical Review Letters.

## 6 PRESENTATION OF TEACHING AND ORGANIZATIONAL ACHIEVEMENTS AS WELL AS ACHIEVEMENTS IN POPULARIZATION OF SCIENCE OR ART

### A. TEACHING EXPERIENCE

- Teaching assistant as an MSc student at Utrecht University for the course Advanced Statistical Physics (2011).
- Teaching assistant as a PhD student at Utrecht University for courses Statistical Field Theory (2012), Quantum Field Theory (2013), and Relativistic and Classical Mechanics (2014).
- Tutorials for the course Statistical Physics A at the University of Warsaw in the academic year 2023-2024 (first year, Master programme).
- Lectures and tutorials for the course Topics in Modern Statistical Physics at the University of Warsaw in the academic year 2023-2024 (first year, Master programme). For this course, I prepared the lecture notes and exercises. The topic was focused on the application of statistical mechanics to soft condensed matter and biophysics.
- Currently, I am supervising a student (Laura Meissner) for her MSc thesis at the University of Warsaw. The project is focused on analytically solving the single-sphere problem in creeping flow for general viscosity tensors.

### B. ORGANIZATIONAL ACTIVITY

- Responsible for evaluating the master programme Theoretical Physics, Utrecht University (2011-2012).
- Member of the education advice committee of the Physics department, Utrecht University (2011-2012).
- Coordinated and divided educational tasks for the Master programme Theoretical Physics, Utrecht University (2014-2016).

### C. SCIENCE POPULARIZATION

- Supervision of a project for high school students about the synthesis and characterization of liquid crystalline materials in 2008, 2009, and 2010 at Eindhoven University of Technology.
- Development of study material for a primary school (“De Hovenier”, Montfort, the Netherlands). The goal was to familiarize primary school students early on with science and technology.
- Organizer of the PLaneT “PhD Lunch and non-expert Talk” seminar, Utrecht University (2015-2016).
- Participation in the European Researcher’s Night Ljubljana to popularize science (2019).
- Participation in the event “Physics in Ljubljana” (2023), an event to popularize scientific research to bachelor students. I contributed by giving a talk on Brownian motion in complex liquids.
- The publication on the Brazil-nut effect in charged colloids had several press releases:
  - Defying gravity: <https://en.uw.edu.pl/defying-gravity/>.
  - First ever observation of Brazil nut effect for colloidal particles: <https://www.uu.nl/en/news/first-ever-observation-of-brazil-nut-effect-for-colloidal-particles> .



## 7 OTHER PROFESSIONAL INFORMATION

- Award “Young Scientists of the IPC PAS (2020)” from the Institute of Physical Chemistry, Warsaw, Poland, based on publication list as the first or corresponding author with IPC PAS affiliation.
- Individual Fellowship from the “Excellence Initiative - Research University” programme, 2022-2024, Institute of Advanced Studies, Warsaw, Poland.
- Award for Outstanding Young Scientist from the Ministry of Education and Science (2022), Poland - the most prestigious stipend for scientists under 35 in Poland.
- Poster prize (first place) at the Departments Day (2015) of Utrecht University, the Netherlands.

## References

- [1] P.-G. De Gennes and J. Prost, *The physics of liquid crystals*, Vol. 83 (Oxford University Press, 1993).
- [2] J. L. Ericksen, *Trans. Soc. Rheol.* **5**, 23 (1961).
- [3] F. M. Leslie, *Arch. Ration. Mech. Anal.* **28**, 265 (1968).
- [4] O. Parodi, *J. Phys. France* **31**, 581 (1970).
- [5] C. W. Oseen, *Trans. Faraday Soc.* **29**, 883 (1933).
- [6] F. C. Frank, *Discuss. Faraday Soc.* **25**, 19 (1958).
- [7] E. Frezza, A. Ferrarini, H. B. Kolli, A. Giacometti, and G. Cinacchi, *J. Chem. Phys.* **138**, 164906 (2013).
- [8] M. Chiappini, T. Drwenski, R. van Roij, and M. Dijkstra, *Phys. Rev. Lett.* **123**, 068001 (2019).
- [9] T. Kato, J. Uchida, T. Ichikawa, and T. Sakamoto, *Angew. Chem. Int. Ed* **57**, 4355 (2018).
- [10] S. T. L. K. Sarp and B. Stebler, *Mol. Cryst. Liq. Cryst.* **60**, 215 (1980).
- [11] R. Oldenbourg, X. Wen, R. B. Meyer, and D. L. D. Caspar, *Phys. Rev. Lett.* **61**, 1851 (1988).
- [12] Y. I. Yaman, E. Demir, R. Vetter, and A. Kocabas, *Nat. Commun.* **10**, 2285 (2019).
- [13] K. Kočevár and I. Muševič, *Phys. Rev. E* **64**, 051711 (2001).
- [14] R. R. Shah and N. L. Abbott, *Langmuir* **19**, 275 (2003).
- [15] M. Raicopol, C. Dascalu, R. Atasiei, and A. T. Ionescu, *J. Appl. Phys* **109**, 124905 (2011).
- [16] D. C. Grahame, *Chem. Rev.* **41**, 441 (1947).
- [17] C. Valeriani, P. J. Camp, J. W. Zwanikken, R. van Roij, and M. Dijkstra, *Soft Matter* **6**, 2793 (2010).
- [18] J.-P. Hansen and I. R. McDonald, *Theory of simple liquids* (Elsevier, 1990).
- [19] V. Fréedericksz and A. Repiewa, *Z. Phys.* **42**, 532 (1927).
- [20] M. Schadt, *Annu. Rev. Mater. Res.* **27**, 305 (1997).
- [21] R. B. Meyer, *Phys. Rev. Lett.* **22**, 918 (1969).
- [22] A. Alexe-Ionescu, *Phys. Lett. A* **180**, 456 (1993).
- [23] M. Gros, S. Gromb, and C. Gavach, *J. Electroanal. Chem* **89**, 29 (1978).
- [24] T. Kakiuchi, *Anal. Chem.* **68**, 3658 (1996).
- [25] H. Jensen, V. Devaud, J. Josserand, and H. H. Girault, *J. Electroanal. Chem* **537**, 77 (2002).
- [26] P. Debye and K. Kleboth, *J. Chem. Phys.* **42**, 3155 (1965).
- [27] A. Onuki, *Phys. Rev. E* **73**, 021506 (2006).
- [28] S. Samin and Y. Tsori, *J. Chem. Phys.* **136**, 154908 (2012).
- [29] M. Bier, A. Gambassi, and S. Dietrich, *J. Chem. Phys.* **137**, 034504 (2012).
- [30] J. D. Jackson, *Classical electrodynamics* (John Wiley & Sons, 2021).
- [31] S. R. De Groot and P. Mazur, *Non-equilibrium thermodynamics* (Courier Corporation, 2013).
- [32] O. D. Lavrentovich, I. Lazo, and O. P. Pishnyak, *Nature* **467**, 947 (2010).
- [33] M. Rajabi, T. Turiv, B.-X. Li, H. Baza, D. Golovaty, and O. D. Lavrentovich, *Phys. Rev. Lett.* **132**, 158102 (2024).
- [34] P. Salamon, N. Éber, A. Krekhov, and A. Buka, *Phys. Rev. E* **87**, 032505 (2013).

- [35] L. Onsager, Phys. Rev. **37**, 405 (1931).
- [36] O. M. Tovkach, C. Conklin, M. C. Calderer, D. Golovaty, O. D. Lavrentovich, J. Viñals, and N. J. Walkington, Phys. Rev. Fluids **2**, 053302 (2017).
- [37] M. Z. Bazant, K. Thornton, and A. Ajdari, Phys. Rev. E **70**, 021506 (2004).
- [38] B. Jerome, Rep. Prog. Phys. **54**, 391 (1991).
- [39] G. P. Crawford, R. Ondris-Crawford, S. Žumer, and J. W. Doane, Phys. Rev. Lett. **70**, 1838 (1993).
- [40] H. Stark, Phys. Rep. **351**, 387 (2001).
- [41] J. Patel and H. Yokoyama, Nature **362**, 525 (1993).
- [42] M. K. McCamley, G. P. Crawford, M. Ravnik, S. Žumer, A. W. Arntstein, and S. M. Opal, App. Phys. Lett. **91**, 141916 (2007).
- [43] V. M. O. Batista, M. L. Blow, and M. M. Telo da Gama, Soft Matter **11**, 4674 (2015).
- [44] A. Rapini and M. Papoular, J. Phys. Colloques **30**, C4 (1969).
- [45] B. W. Ninham and V. Parsegian, J. Theor. Biol. **31**, 405 (1971).
- [46] D. G. Rowan, J.-P. Hansen, and E. Trizac, Mol. Phys. **98**, 1369 (2000).
- [47] E. Trizac, L. Bocquet, R. Agra, J.-J. Weis, and M. Aubouy, J. Phys.: Cond. Matt. **14**, 9339 (2002).
- [48] C. Álvarez and G. Téllez, J. Chem. Phys. **133**, 144908 (2010).
- [49] J. C. Everts, Phys. Rev. Res. **2**, 033144 (2020).
- [50] B. Derjaguin and L. Landau, Acta Physicochim. URSS **14**, 633 (1941).
- [51] E. J. W. Verwey and J. T. G. Overbeek, in *Theory of the Stability of Lyophobic Colloids* (Elsevier, New York, 1948).
- [52] N. D. Mermin, Rev. Mod. Phys. **51**, 591 (1979).
- [53] G. P. Alexander, B. G.-g. Chen, E. A. Matsumoto, and R. D. Kamien, Rev. Mod. Phys. **84**, 497 (2012).
- [54] J. J. Sandford O'Neill, P. S. Salter, M. J. Booth, S. J. Elston, and S. M. Morris, Nat. Commun. **11**, 2203 (2020).
- [55] A. A. Verhoeff, R. H. J. Otten, P. van der Schoot, and H. N. W. Lekkerkerker, J. Chem. Phys. **134**, 044904 (2011).
- [56] M. Nikkhou, M. Škarabot, S. Čopar, M. Ravnik, S. Žumer, and I. Muševič, Nat. Phys. **11**, 183 (2015).
- [57] S. Čopar, Ž. Kos, T. Emeršič, and U. Tkalec, Nat. Commun. **11**, 59 (2020).
- [58] B.-X. Li, V. Borshch, R.-L. Xiao, S. Paladugu, T. Turiv, S. V. Shiyankovskii, and O. D. Lavrentovich, Nat. Commun **9**, 1 (2018).
- [59] J. Pollard, G. Posnjak, S. Čopar, I. Muševič, and G. P. Alexander, Phys. Rev. X **9**, 021004 (2019).
- [60] I. I. Smalyukh, Rep. Prog. Phys **83**, 106601 (2020).
- [61] A. Yethiraj and A. van Blaaderen, Nature **421**, 513 (2003).
- [62] M. Westbroek, N. Boon, and R. van Roij, Phys. Chem. Chem. Phys. **17**, 25100 (2015).
- [63] N. Schopohl and T. Sluckin, J. Phys. France **49**, 1097 (1988).
- [64] N. Schopohl and T. J. Sluckin, Phys. Rev. Lett. **59**, 2582 (1987).
- [65] S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnár, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, Science **294**, 1488 (2001).
- [66] G. Müller and J. Senior, J. Hydraul. Res. **47**, 666 (2009).
- [67] X. Tang and J. V. Selinger, Soft Matter **15**, 587 (2019).
- [68] X. Chen, E. Korblova, D. Dong, X. Wei, R. Shao, L. Radzihovsky, M. A. Glaser, J. E. MacLennan, D. Bedrov, D. M. Walba, and N. A. Clark, Proc. Natl. Acad. Sci. U.S.A. **117**, 14021 (2020).
- [69] M. U. Khan, J. Kim, M. Y. Chougale, R. A. Shaikat, Q. M. Saqib, S. R. Patil, B. Mohammad, and J. Bae, Adv. Intell. Syst. **5**, 2200281 (2023).
- [70] S. Dai, X. Liu, Y. Liu, Y. Xu, J. Zhang, Y. Wu, P. Cheng, L. Xiong, and J. Huang, Adv. Mater. **35**, 2300329 (2023).
- [71] N. Kavokine, M.-L. Bocquet, and L. Bocquet, Nature **602**, 84 (2022).
- [72] R. P. Thedford, F. Yu, W. R. T. Tait, K. Shastri, F. Monticone, and U. Wiesner, Adv. Mat. **35**, 2203908 (2023).
- [73] V. Sultanov, A. Kavčič, E. Kokkinakis, N. Sebastián, M. V. Chekhova, and M. Humar, Nature **631**, 294 (2024).