Final report

on the project entitled

Tunable topology of confined soft matter

(NKFIH FK 125134)

The research performed in the project NKFIH FK 125134 targeted mainly the comprehensive exploration and understanding of phenomena related to the topology of anisotropic soft matter tunable by external stimuli. In the five years of the project, we published 31 papers including the acknowledgement of the project with cumulated impact factor: 141.097. We already obtained 136 independent citations to these papers published recently. There is one additional manuscript in peer review, and three manuscripts in preparation.

1. Topology of liquid crystals in spherical cap shaped confinement

According to the workplan, and in connection with all work packages: WP1 - Director alignment at liquid crystal interfaces with non-solid media, WP2 - Topology in frustrated anisotropic soft matter, and WP3 - Control of topological structures by external fields, a major part of our results was related to sessile droplets. The connection between the results and the tasks in the original workplan is indicated in brackets with the letter T and the task number.

We measured the orientation of several nematic liquid crystals at interfaces with air and the corresponding director structure and topology in sessile droplets [1] (T1.2, T1.3, Fig.1a). We identified two types of temperature triggered orientational transitions, one on the coated substrate and another at the free surface bounded by air. We revealed two distinct textures involving topological defects governed by the reorganization of the director field, moreover, we detected a double transition, involving a re-entrant behavior.

We studied the sessile nematic droplets in magnetic/electric fields and light, because such objects exhibit non-trivial topological properties and have potential for application in tunable multifocal optical lenses. First, in order to understand the director structure and its tunability, we applied magnetic fields on the droplets [2] (T3.2, Fig.1b). We showed that at sufficiently high horizontal magnetic fields, a Néel wall-type metastable inversion wall forms in the middle of the drop, which moves outward. Applying fields above a critical angle between the plate of the spherical cap and the magnetic field, a uniform director structure emerges. Drops with uniform director structure can be used as tunable optical lenses, where the focal length can be controlled by light polarization, viewing angle, and external fields. We proposed a theory that explains the texture variation in small magnetic fields and accounts for the formation and motion of the inversion wall. We studied the droplets also in electric fields [3]. Similar to our findings in DC magnetic fields, we found the formation of an inversion wall normal to AC electric fields. While at low frequencies, the direction of the wall is stationary, at higher frequencies it turns toward the external electric field. In both cases, the defect wall is also swept toward the periphery of the drop, where it eventually disappears. The linear displacement of the electric field-induced defect wall could be described by our theoretical description.

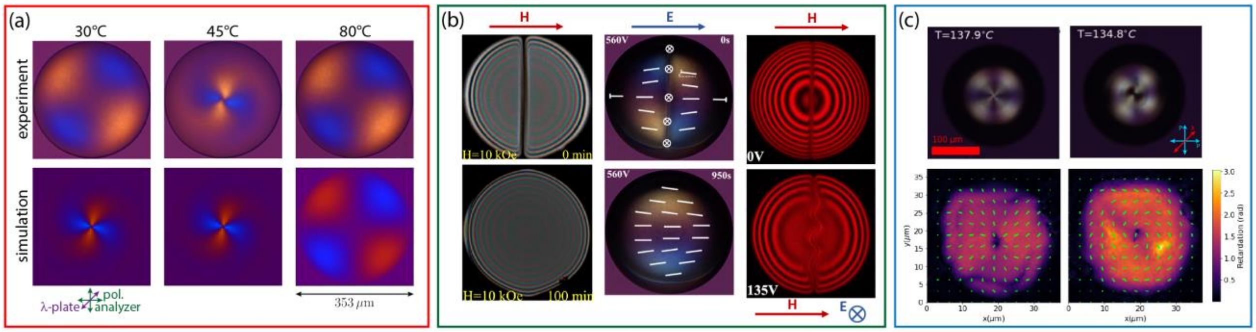

Figure 1: Polarizing microscopy textures of liquid crystal droplets with different structure and topology. (a) Anchoring transition at the air interface [1]. (b) Effect of magnetic and electric fields [2-4]. (c) Ferroelectric nematic droplet [6].

The rotation of the defect wall at high frequencies is a result of the antiparallel orientation of the effective moment vector and the electric field due to the lower dielectric constant and higher electric conductivity of the defect wall than of those of the rest of the liquid crystal droplet. We continued our studies on droplets subjected to simultaneous competing magnetic and electric fields. It was found that above a threshold electric field, the initially straight defect wall buckles to form a zig-zag shape [4].

We investigated the topological structure of sessile droplets of a photo-responsive dimer exhibiting the transition between nematic and twist-bend nematic phases [5] (T1.3, T2.2). The internal director structures of these droplets varied depending on the phase and on the imposed boundary conditions. The switching between the nematic phases was successfully performed either by temperature control or by UV light irradiation. The director field in the droplets was found to be substantially different from the case of conventional nematics due to the extremely small bend elastic constant and the planar alignment at the air interface.

We studied sessile droplets of a liquid crystal exhibiting a classical nematic and a new type of ferroelectric nematic phase [6] (Fig.1c). We discovered and explained a new phenomenon, namely a thermal gradient-induced circular flow in the drops around topological defects. We provided a simple model showing that the tangential arrangement of the ferroelectric polarization combined with the vertical thermal gradient and the pyroelectricity of the fluid drives the rotation. The director structure of the droplets in the ferroelectric phase was found to be skyrmion-like.

We broadened our studies on liquid crystal droplets to a composite system of liquid crystals and nanoparticles (T1.2, T1.3, T2.3). We demonstrated experimentally that the anchoring of a nematic liquid crystal on a solid substrate together with the anchoring of the liquid crystal on a nanoparticle surface induces orientational self-assembly of anisometric nanoparticles in liquid crystal droplets. The observed phenomenon opens a novel route for fabrication of thin colloidal films with tailored properties [7].

2. Topology and patterns of liquid crystals in planar confinement

We investigated a nematic liquid crystal in sandwich cells showing a stable lattice of topological defects induced by electric field (T3.2). We demonstrated the generation of optical vortices by individual topological defects and by diffraction on dislocations of the lattice structure. In both cases, the vortex-generation efficiency was tunable by the applied voltage [8–10] (Fig.2a).

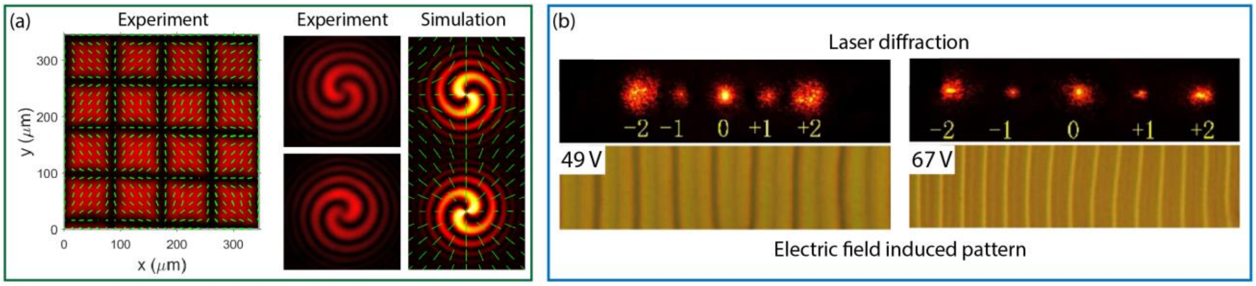

Figure 2: (a) The structure of the electric field induced defect grid measured by polarimetric microscopy, and interferograms confirming optical vortices [8]. (b) Tunable electric field induced patterns used as laser deflectors [12].

We demonstrated that large-scale patterning of topological defects is possible in anisotropic fluids despite their unfavorably high-energy states [11] (T1.4, T3.2). We presented a pathway for topology engineering: processing topological defects and shape large-scale patterns in materials with liquid crystalline nature. Using either electric-field driven temperature gradients or material flow, we created dragging fields acting on the interface between liquid crystalline and isotropic phases. The specific alignment of liquid crystal interfacing with an isotropic fluid and the dragging resulted in large-scale patterns of topological defects.

We studied unusual electric field induced pattern formation in a newly synthesized hockey-stick nematic liquid crystal [12] (Fig.2b). Extraordinary pattern morphologies were found showing rich variety as a function of frequency, temperature, and initial director alignment. The behavior was compared with those observed in rod-like and bent-core nematics. The extraordinary findings were attributed to the cybotactic nematic phase including polar smectic clusters, with the influence of flexoelectricity. Our investigations of flexoelectric domains for several nematic liquid crystals demonstrated that the voltage dependent wavenumber of the periodic pattern structure varies almost linearly with the voltage above a threshold [13]. This was confirmed by a numerical analysis of the full nonlinear equations for the director field and the induced electric potential.

Our work [14] (T3.1, T3.2) revealed the influence of UV light-induced pitch variation on electroconvection patterns of a chiral nematic liquid crystal containing a photo-responsive chiral dopant. The ability to control the orientation of convection rolls by tuning the light intensity can be used as a switchable optical grating. We performed experiments on a new photo-responsive bent-core nematic material, which exhibits flexoelectric domains [15]. We found that the pattern morphologies, electric threshold, and periodicity can be controlled by light, which effect originates in the photoisomerization within the molecules. Based on this principle, a prototype of controllable optical grating was assembled, whose operation can be manipulated by the wavelength or the intensity of light. Due to the easy, instant, and remote operation by light, this optical, contactless tunability has a great advantage over traditional electric control in tunable photonic devices. Electroconvection based patterns were reported in a hybrid aligned cholesteric liquid crystal containing a chiral photosensitive dopant [16] (T3.4). We showed that gratings induced by a periodic flow provide wide tunability utilizing the pattern formation mechanism and its dependence on the material properties. The irradiation by UV light rotated the pattern via alteration of the helical pitch, where the magnitude and direction of the rotation angle depended on the UV light intensity. A prototype grating capable of two-dimensional beam steering was demonstrated.

We investigated the generation of twist deformation in nematic liquid crystals using photoalignment [17] (T3.1). By gradually increasing the twist angle, supertwisted cells were constructed in the range of 2 pi – 3 pi twist angle. The supertwist relaxed through the formation of topological defects, depending on the character of the photosensitive substrate. The difference in the relaxation process can be related to the zenithal anchoring strength on the photosensitive substrate. We provided experimental evidence that photoalignment at the nematic liquid crystal - polymer interface cannot be simply considered as a two-dimensional process [18,19] (T3.1). Our experiments clearly indicated that the photoaligning process does not depend on the individual properties of the materials involved. The polymer-liquid crystal interface should be regarded as a coupled system, where the two components mutually influence each other. Substantial differences have been found in the photo-alignment process depending on the molecular structure [20]. Moreover, we found that the efficiency of photoalignment exhibits marked differences depending on the structure of the rigid core of the liquid crystal molecules [21]. It was demonstrated that the photo-orientation process is also significantly affected by the type of mesophase in which irradiation is carried out.

3. Supplementary results

In accordance with the workplan of the project, additional to the specified tasks, we performed measurements of material parameters/properties of the studied compounds. In this chapter, we summarize our results related to such supplementary tasks.

We characterized the viscoelastic properties of the liquid crystal dimer used in the studies related to droplets [5] and found extraordinary behavior [22,23]. We showed that the dimer exhibits photoswitching of its viscoelastic properties (shear viscosity, storage, and loss moduli) with remarkable contrast of up to 1:1000000 while transitioning between crystal and nematic phases. The combination of highly contrasting viscoelastic behavior with fast and reversible switching establishes a whole new performance level for mechanically responsive organic materials and offers very considerable application potential in such diverse areas as photo- switchable adhesives, in vibration control, and as novel brakes. We studied the rheological and rheo-optical properties of suspensions of anisometric pigment particles in a non-polar fluid [24]. Different rheological regimes from the dilute to an orientationally arrested gel state were characterized and compared with existing theoretical models. A unique combination of the optical properties of the particles results in a giant rheo-optical effect: an unprecedentedly large shear stress-induced birefringence was found. We also characterized a new liquid crystal dimer [25]. The spectroscopic properties were analysed by UV–Vis and fluorescence techniques. The obtained results present a solid basis for the future studies on the stilbene-based liquid crystal dimers, thus affording guidelines for development of a structure–property relationship of these compounds. Moreover, we characterized two series of new light sensitive bent-core liquid crystals [26,27]. The results were compared with those reported for the azobenzoyl analogues.

In bent-core and rod-like liquid crystals, in their mixture, and in samples doped with magnetic nanoparticles, we investigated the influence of magnetic field on the isotropic-to-nematic phase transition temperature [28]. A magnetic-field-induced negative or positive shift of the transition temperature was detected depending on the magnetic field orientation with respect to the initial orientation of the nematic phase, and on the type of the liquid crystal. The dynamic magnetic susceptibilities of liquid crystals doped with magnetic nanoparticles were measured [29]. In soft elastomers containing magnetic particles, we characterized the tunability of surface topography in response to magnetic fields [30]. We measured the physical properties including tilt angle and spontaneous polarization of ferroelectric liquid crystalline mixtures based on chiral components [31].

4. Administrative report

P. Salamon participated in the following outreach events giving presentations related to the subject of the project: “Festival of the Hungarian Science” (also interviewed and appeared in the scientific television show “Minden tudás”), “Night of the Researchers” (all years), “Simonyi-nap” (2020), at “ELTE TDK hét” (2020), “Mafihe Téli Iskola” (2021).

M. Máthé joined the project as an MSc. student (Physics, ELTE) in 2019, then as a PhD student (Physics, ELTE) from 2020 under the supervision of P. Salamon. A.R.K. Nassrah joined the project as a PhD student (Physics, ELTE) in 2019 under the supervision of T. Tóth-Katona. Starting from December 2020, the salary of P. Salamon is not financed from the budget of the project. As a result, hiring of a BSc student, Bendegúz Farkas (Physics, ELTE), became possible. The student provided technical assistance related to goals of the project, e.g., he prepares samples, performs experiments. V. Kenderesi joined the project in 2018 as a technician.

Budapest, September 30, 2022

Dr. Péter Salamon

principal investigator

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