Final report

on the project entitled

Interfacial topology of anisotropic soft matter

(NKFIH PD 121019)

The research performed in the project NKFIH PD 121019 targeted mainly the comprehensive exploration and understanding of phenomena related to the topology of anisotropic soft matter interfacing other fluid media including gases and liquids.

The first work package (WP1) of the project was to build and test a new experimental apparatus, which is capable to determine the interfacial molecular orientation (director). This task was completed, and additional functions of the apparatus were also developed, allowed by the financial support of the host institution and the National Research, Development and Innovation Office by granting the project FK 125134 with the lead of the principal investigator (P. Salamon). The goals of the present project (PD 121019) are included entirely in those of FK 125134, but this latter project (started on 1st September, 2017) provides funding for investments and consumables as well.

The device is the main part of the new optical laboratory established in 2017, providing a state-of-the-art facility for the research conducted in the current project. The setup was constructed in a way that allows the fast switching between operation modes without the need of sample movement or re-tuning the optics. Additionally, the apparatus had to be built in a vibration isolated upright position to allow measurements on liquids with free surface. The instrument can be operated in the following modes: (1) transmission geometry, polarizing microscopy with white light illumination, color camera; (2) transmission geometry for fast birefringence mapping: by alternating monochromatic stroboscopic illumination at two wavelengths, two high speed birefringence modulators (Pockels cells), high speed monochrome camera; (3) episcopic mode, using optionally polarized white or monochromatic light; (4) anisotropic reflectivity mapping in episcopic geometry, using two liquid crystal birefringence modulators; (5) modes 1-4 with infrared laser coupling for local sample heating or patterning by a motorized XY stage, (6) modes 1-4 with a UV or violet laser coupling for attenuation of light sensitive samples; (7) direct laser writing with the UV laser by a motorized XY stage for photolithography in order to create patterned microelectrode systems, and microfluidic chips

By using the fast birefringence mapping in mode 2, studying the dynamics of any birefringent material including biological samples or liquid crystals, had become possible at unprecedentedly high speeds up to 2000 frames per seconds at 512x512 pixels resolution. For such specification, the measurement range of the retardation is from 0 to pi in radians. The extension of the this range up to about 5 pi can be done by using alternating illumination at two wavelengths, at the cost of the reduction of speed by a factor of two. We note that the commercially available birefringence mapping instruments can measure retardation maps up to pi phase difference with frame rates lower than 30 Hz.

As also planned in WP1, we successfully developed a software to simulate the light propagating in an arbitrary three dimensional director field. The program can simulate polarizing microscope images with arbitrary polarizer and wave plate settings and it is very useful to test and confirm three dimensional director structures realized in our experiments. The general software applies either Müller matrix or Jones matrix based calculations in a voxelized space, where each voxel is considered as a birefringent element with a slow axis direction and retardation magnitude depending on the considered director field and material parameters. In the Müller matrix mode, we can simulate systems with partially polarized light, while with Jones matrices, besides calculating polarizing microscope images, we can also simulate light diffraction on director structures. Our software is also capable to simulate white light illumination including a number of light wavelengths with proper weighting to have comparable colors found in experiments.

The measurements of the interfacial alignment at nematic-air interfaces were performed in a number of liquid crystals, systematically varying the molecular structure (WP2, WP3). The molecular orientation direction (the director) was found to be perpendicular to the nematic-air interface in case of the most of the materials tested including 5CB, 6CB, 8CB, CCN47, CCN55, ZLI1497, 7OCB, ZLI1695, PCH7, and several others. More interestingly, we found some materials where the director is not perpendicular to the interface, which resulted in completely different topology of director field and thus different looking textures in polarizing microscopy. Such materials include CB6OABOBu, a photo-sensitive dimer, and the liquid crystals D6AB, Phase4, Phase5 containing azoxy bridges.

We compared the molecular orientation fields (director structures) in sessile droplets of the standard liquid crystal 5CB and the novel photosensitive dimer molecule on different surface alignment conditions. We found that the different boundary conditions on the solid substrate and on the interface with air result in distinct topological states. The molecules of 5CB tend to align perpendicular to the free surface, while the interfacial director angle is close to zero for the dimer. In case of the photosensitive material, the realized topological structures can be tuned by light. Our results are related to the milestones M3 and M4, and were summarized in a paper [1]. Other manuscripts in this topic are in preparation.

As a supplementary task, we characterized the rheological properties of the same photosensitive dimer. We showed that CB6OABOBu exhibits photoswitching of its viscoelastic properties (shear viscosity, storage and loss moduli) with remarkable contrast of up to one to a million while transitioning between crystal and nematic phases. This switching is reversible and takes less than 100 s for both forward and backward reactions due to the coexistence of two allotropes containing two types of stereoisomers. This 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 novel brakes, in vibration control, and as photoswitchable adhesives. We published our results in this matter in two papers [2,3].

We also studied the surface topographical modifications of a soft magnetoactive elastomer in response to magnetic fields. Optical profilometry analysis showed that the magnetic field-induced surface roughness is in the range of 1 mm/T. Shape analysis of sessile water droplets deposited on the magnetoactive surface revealed that the contact angle of water can be changed by magnetic field in a reversible way. Despite the increased surface roughness, the apparent contact angle decreased with increasing field, which was attributed to the field-induced protrusion of hydrophilic microparticles from the surface layer. The above results were published in one paper [4].

As a supplementary task, we characterized new multicomponent liquid crystal mixtures exhibiting ferroelectric smectic phase at room temperature (one paper published [5]). These materials showed high spontaneous polarization, above 100 nC/cm^2, and short helical pitch, below 180 nm, making them good candidates for applications using deformed helix ferroelectric mode in photonics and opto-electronics. We plan to study the interfacial topology of such ferroelectric liquid crystals, because their director alignment at free surfaces are unknown, and more importantly their high sensitivity to electric fields promises the presence of unprecedented, interesting effects like electrically controllable superwetting/spreading. Those studies could, however, not be started yet, due to the time limitation of the present project.

We studied a nematic liquid crystal showing a stable lattice of topological defects induced by electric field in order to generate optical vortices appearing as donut beams due to their singularity in their phase. Optical vortices are important because of their already existing uses (e.g. in super-resolution microscopy or in laser tweezers), and due to their promising emerging applications (e.g. in optical communication). This part of our work was closely related to WP7 and milestone M5, thought the studied system was not bounded by a fluid but solid surfaces. We performed experiments including quantitative polarimetry and laser diffraction. We demonstrated the generation of optical vortices by the topological defects in the case of two distinct mechanisms. First, individual defects converted circularly polarized light partially into a vortex beam with opposite handedness, while beams diffracted on the defect lattice did not carry vorticity. Second, an edge dislocation of the lattice structure was a topological defect on a larger length scale; then beams diffracted on a single dislocation possessed optical vortex character. The vortex-generation efficiency was tunable by the applied voltage and it was found to be close to 100% even in the case of light with different wavelengths. We supported our experiments by simulations using our Jones matrix based software mentioned above. Our results were published in two papers in this topic [6,7]. A third manuscript is submitted now [8]. The work related to optical vortices was shown to the public in oral presentations by P. Salamon in the following events: 27th International Liquid Crystal Conference, Kyoto, Japan (22nd-27th July, 2018); Eutopia-2: Second meeting of the European Topology Interdisciplinary Initiative, San Sebastian, Spain (4th-7th November, 2019); public seminar at Wigner Research Centre for Physics, Budapest, Hungary (19th February, 2019); public talk at the Hungarian Academy of Sciences, Budapest, Hungary (10th May, 2019); Hungarian Physicist Meeting, Sopron, Hungary (22nd August, 2019).

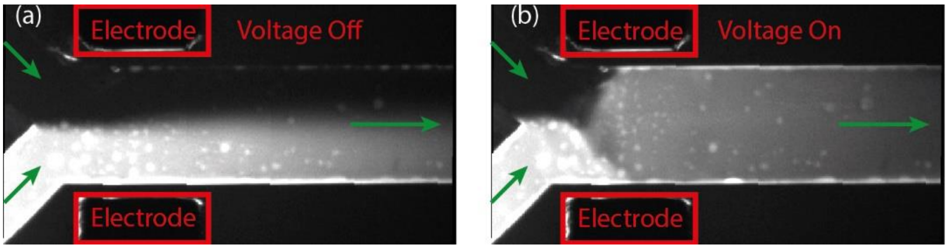

Figure 1: Observation the mixing of two identical liquid crystals in a microfluidic Y-junction by fluorescent microscopy. The lower branch is doped with a fluorescent dye. (a) Mixing by diffusion. (b) Mixing by electroconvection. The width and depth of the merged channel was 100 µm and 10 µm, respectively.

Also, in relation with milestone M5 and WP7, we investigated electric field induced flow vortices in confined nematic liquid crystals, however, the confinement was not achieved by gases of liquids, but by solid interfaces. Nematic liquid crystals with negative dielectric and positive conductivity anisotropies become unstable above a critical voltage and ordered flow vortices appear as roll structures in one dimensional confinement (sandwiched between two glass plates). Our investigations showed that such vortices may exist in different geometries of confinement as well; their presence was confirmed also in microfluidic channels. In flows of isotropic liquids in microfluidic channels, it is difficult to observe turbulent vortices, since the system is typically characterized by low Reynolds numbers. As a consequence, mixing of fluids in laminar flow is slowly achieved by diffusion. Nematics, however, offer an electrically adjustable way to generate vortices with flow velocities comparable to typical flow speeds applied in microfluidics. We showed the characteristics of electric field induced vortex formation (vorticity, flow field) and present its application in microfluidic mixing. We revealed that complete mixing of fluids after a microfluidic Y-junction can be achieved by electroconvective flow vortices in significantly smaller length compared to the usual diffusive mixing generally used in microfluidics (Figure 1). This work was presented in a poster at the 27th International Liquid Crystal

Conference, Kyoto, Japan (22^{nd}-27^{th} July, 2018) [9]. A manuscript based on this matter is before submission.

We extended our investigations of interfacial director alignment to interfaces where the liquid crystal is bounded by a liquid as planned in WP5. We studied several types of liquid crystals including 5CB and E7, bounded by their own isotropic fluid phase and found that the interfacial director is at an angle with respect to the interface. When the isotropic-nematic interface was moving due to a temporally changing temperature field, we found an extraordinary, yet undiscovered type of pattern forming phenomena that results in a large-scale array of topological defects that are in a high-energy state but still stationarily stable due to topological protection (Figure 2). We clarified that the pattern formation is rooted in collective long-range repulsive interactions between topological defects with identical topological charges formed at the isotropic-nematic interface due to the tilted orientation. The strength of the dragging force and surface interactions are found to be dominant factors in generating and stabilizing the patterns. The discovered patterning method endows fluids with tailored topological patterns at desired positions, paving a new way for the development of fluids and gels with spatially modulated topological nature, which is vital and challenging from both fundamental and technological points of view in anisotropic fluids. We submitted a manuscript about this work to Soft Matter [10].

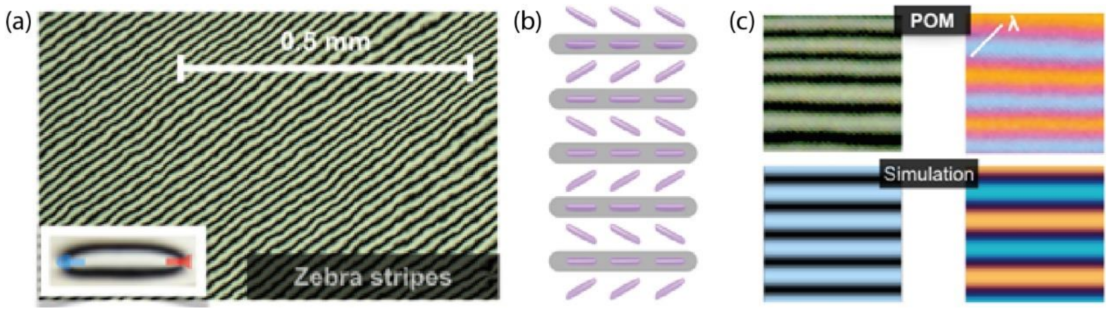

Figure 2: (a) Large scale topological pattern observed in polarized optical microscopy (POM); inset: the two ends of an elongated stripe are of one-half topological strength defects with opposite topological charge. (b) A schematic of the director field of the stripes. (c) A detailed POM view and a simulated image of the Zebra stripes with crossed polarizers, and with a λ-plate inserted. The simulation is made by the Jones matrix method.

As part of WP7 and in relation with milestone M5, we performed detailed experimental and theoretical studies of the director structure of a liquid crystal (8CB) in spherical cap shape in the presence of an external magnetic field (B). The droplet was sitting on a solid substrate, while its curved surface was in air with perpendicular interfacial director orientation. We applied magnetic fields of various amplitudes and directions with possible practical applications in mind (Figure 3). We described the magnetic field-dependent director structure experimentally and theoretically. We showed that above a threshold field, a Néel wall-type metastable inversion wall forms in the middle of the drop (Figure 3b) and then moves outward (Figure 3c). The wall does not appear above a critical angle between the plate of the spherical cap and the magnetic field, thus giving rise to uniform director structure. After polymerization, such objects can be used to make optical lenses with focal length variable with the polarization and propagation direction of the light. We already prepared a manuscript in this topic, which is before submission [11].

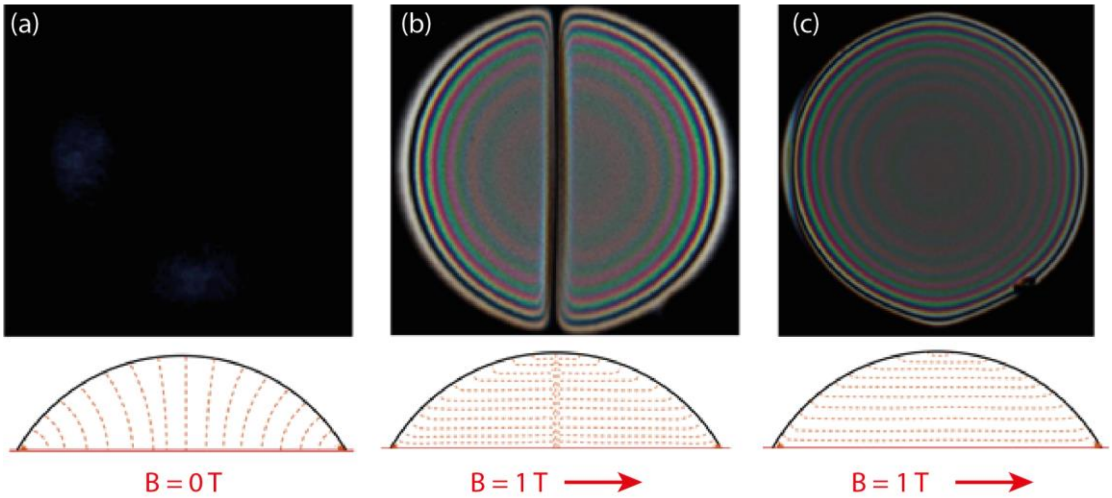

Figure 3: Top view of textures between crossed polarizers at ±45° with respect to the magnetic field (horizontal) of a 490 µm diameter liquid crystal drop illuminated by white light, and the corresponding director structure in side view, with (a) no magnetic field, (b) B = 1 T with an inversion wall in the middle, and (c) B = 1 T with the inversion wall moved to the left side.

As described above, all work packages and milestones of the project have been addressed, except WP6 and M7 that are related to embedding particles in liquid crystals. The work on this has actually been started recently; we got some promising results, but those are not public yet. Our first aim with the particles was to investigate their interaction and manipulation with the magnetic field induced, movable inversion wall described above. In the first experiments, we used polystyrene microspheres that exhibit parallel director alignment.

In summary, we published 7 papers [1-7], - including acknowledgement of the project -, with cumulated impact factor of 23.831. We already obtained 20 independent citations to these 7 papers published recently. There are two additional manuscripts [8,10] in peer review, and two manuscripts are before submission [9,11]. The results of the project were presented in 5 oral and 1 poster presentations. P. Salamon also participated in outreach activities giving presentations related to subject of the project: in the Festival of the Hungarian Science at the Wigner Research Centre for Physics for groups of high school students (also interviewed and appeared in the scientific television show “Minden tudás”), and in the Night of the Researchers. P. Salamon hosted two high school students in a one-week research practice programme, where one task was to observe liquid crystal droplets with different director structures. In the fall of 2019, P. Salamon has become the supervisor of a physicist MSc. student (M. Máthé from Eötvös Loránd University) whose tasks include the investigation of topological transitions in liquid crystal droplets, which is strongly related to the topic of the project under scope.

Budapest, December 23, 2019

Dr. Péter Salamon

principal investigator

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