Review

Interactions, topology and photonic properties of liquid crystal colloids and dispersions

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Received 29 June 2018 / Received in final form 20 September 2018 Published online 28 March 2019

Abstract. Liquid crystal-based colloids demonstrate new type of colloidal interaction between the particles, which is of elastic origin, has very long range and is extremely strong, with pair-binding elastic potential exceeding several thousands of $k_{\rm B}T$. This gives rise to novel and strong colloidal assembly mechanisms, which can be used to assemble 2D and 3D colloidal crystals. This mini-review discusses colloidal pair interaction mechanisms and topological aspects of colloidal interaction. Topological defects, which are responsible for elastic forces between colloidal particles show an amazing diversity of different structures, including colloidal entanglement, knotting and linking of particles. An introduction is given to dispersions of liquid crystal droplets in an immiscible fluid, which represent a new class of photonic micro-devices, including micro-lasers, micro-fibers and optical micro-cavities made of tuneable liquid crystals.

1 Introduction

The first indication of forces between particles which are included in the nematic liquid crystals was given in X-ray scattering experiments on a lyotropic nematic crystal by Raghunathan et al. [1]. In those experiments, nanoparticles were dispersed in the lyotropic liquid crystals and the scattering data provided indications of the assembly of nanoparticles in a liquid crystal. Whereas this experiment gave an indication of attractive forces between small particles in liquid crystals, the first direct proof of particle attraction in the nematic liquid crystals was given in the experiment by Poulin et al. [2]. They used a dispersion of water droplets in the nematic liquid crystal and observed clear attraction between micro-meter size droplets. The droplets spontaneously assembled into chains of droplets, separated by topological defects. These topological defects are formed spontaneously whenever an inclusion is inserted in the nematic liquid crystal and the liquid crystal aligns all over the closed surface of the inclusion.

It turns out that topological defects around inclusions in liquid crystals have the major role in generating forces between colloidal inclusions [3-6]. Due to the fact that liquid crystal molecules are forced to align along a closed surface of the inclusions,

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the liquid crystal is strongly elastically deformed around the inclusion. When two such particles are brought together, the regions of distortions around each particle start to overlap. Consequently, the free energy of a pair of particles depends on their separation, resulting in a structural force between the two particles which is therefore of elastic origin. Actually, one of the first observation of attractive structural forces between particles in nematic liquid crystals has been realized by Cladis et al and Rault et al. in the 1970s in experiments, where particles were used to "decorate" the director on a free surface of the nematic liquid crystal [7,8].

The forces between colloidal inclusions have been analysed theoretically [9–21] and measured experimentally [22–34] by many groups. They are classified into two main categories, depending on the type of topological defect accompanying the particle and the spatial symmetry of interaction. We shall consider only perpendicular orientation of liquid crystal on the surface of the colloidal inclusion. In this case, the first type is dipolar colloidal particle, where each particle is accompanied by hyperbolic point defect, also called the hyperbolic hedgehog. The corresponding pair interaction force between two dipolar colloidal particles is of long range, exceeding tens of micrometers for micrometer-sized particles. The corresponding pair interaction energies are of the order of several 1000 $k_{\rm B}T$. The other type is of quadruplar symmetry and the colloids are called quadrupolar colloids. The colloidal particle is encircled by a closed defect ring [9,35], also called the Saturn ring. The pair interaction forces of two quadrupolar nematic colloids are much lower in magnitude compared to dipolar forces. Their pair interaction energy is nearly an order magnitude lower compared to dipolar colloids and is of the order of 100 $k_{\rm B}T$.

Whether the colloidal inclusion appears as a dipolar or quadrupolar, colloidal particle depends on its size and the geometric shape, the strength of the orientational anchoring of liquid crystal molecules on the particle surface, confinement and external fields [36]. In most cases, the inclusion is of spherical shape, which allows for precise experimental analysis of the orientation field around the colloidal inclusion and measurements of pair interaction forces between spherical microspheres in liquid crystals. For a given strength of surface anchoring of liquid crystal molecules, it was demonstrated both experimentally and theoretically that the bigger microspheres usually adopt a dipolar configuration. By reducing the diameter of the microsphere, this dipolar configuration is transformed into a quadrupolar configuration. By further reducing the size of the microsphere, the required elastic deformation of the liquid crystal around the particle becomes so large that the orientational anchoring at the surface is no longer efficient in maintaining perpendicular surface anchoring. The characteristic length which describes this transitional regime is the surface extrapolation length [37]. It is defined as the ratio of the elastic constant of the liquid crystal and the surface anchoring energy coefficient:

$$\xi_s = \frac{K}{W_a}.\tag{1}$$

Here W_a is the anchoring energy coefficient describing the strength of surface anchoring of liquid crystal, whereas K is the elastic constant of the liquid crystal. For a weak surface anchoring ($W_a \sim 10^{-6} \text{ J/m}^2$) and the typical elastic constant $K \sim 10^{-11}$ N, the extrapolation length is around $\xi \sim 10 \,\mu\text{m}$. For very strong surface anchoring ($W_a \sim 10^{-3} \text{ J/m}^2$), the extrapolation length decreases to around $\sim 100 \,\text{nm}$, but is still much larger than the typical liquid crystal molecules (several nm). This means that very small particles in the nanometre range will be inefficient in imposing elastic deformation and the orientation around the particle will be uniform. As a result, the particle will be "invisible" to other particles floating in the liquid crystal and there will be no interaction of elastic origin between them. Majority of studies of nematic colloids have been performed on micrometer-sized colloidal particles in nematic liquid crystals, and the particles were usually of spherical shape. The reason is widespread availability of well defined spherical particles, which are used in colloidal chemistry and physics. The aim of these studies is to study novel colloidal interactions on the micrometer-scale, as well as their extension to the nanoscale, where the tools of observation and manipulation of nanoparticles are very limited.

On the other hand, studies of nanoparticle dispersions in the nematic liquid crystals aimed at changing the material properties of such a nanocomposite [38-53]. For example, by taking quantum dots and dispersing them in the nematic liquid crystal one aims at obtaining a uniform dispersion of quantum dots in the nematic liquid crystal, serving as a uniform fluorescent medium for photonic applications. By taking ferroelectric nanoparticles and dispersing them in the nematic liquid crystals one aims at obtaining ferroelectric nematic liquid crystal, where the quadrupolar symmetry of the nematic ordering field is coupled with the vector field describing macroscopic ferroelectric polarization. In another example, magnetic nanoparticles dispersed in the nematic liquid crystal could provide microscopic ferromagnetic ordering in such a composite material. After more than a decade of work, ferromagnetic crystals were successfully realized in the dispersion of solid ferromagnetic nano-platelets in the nematic liquid crystal [54]. Quite recently, a hybrid nematic material showing optical biaxiality was discoverd [55]. In this system, two different fluid systems are interconnected, i.e. the nematic host and the ordered fluid of inorganic colloidal rods. This successful realization of an efficient coupling between nanoparticle properties and the microscopic alignment of liquid crystal can undoubtedly present a guideline for realizing ferroelectricity and uniform fluorescence in nematic liquid crystals doped with nanoparticles.

This article is a short review of underlying physical phenomena in dispersions of particles of various sizes and shapes in the nematic liquid crystals. We shall discuss the interaction of microparticles in the nematic liquid crystals, covering dipolar and quadrupolar colloids and we shall make a brief excursion into the more complex binding of colloidal particles, which is called colloidal entanglement. The primary purpose is to give a clear insight into the size and shape-dependence of pair colloidal interactions. A part of this article is devoted to topological aspects of nematic colloids, where we aim at basic level of understanding of what is observed in the experiment. As a curiosity, photonic properties of "inverted" colloidal systems, where the "particles" are liquid crystal droplets dispersed in an immiscible fluid, are introduced in the last section.

2 Interaction of spherical particles in the nematic liquid crystals

We discuss nematic liquid crystal dispersions of spherical particles with perpendicular surface anchoring of liquid crystal molecules. The reason we choose such a geometry is huge variety of topological phenomena, which are observed for this type of surface anchoring. It turns out that the other type of surface alignment, i.e. the planar surface anchoring, gives rise to surface topological defects, which cannot be manipulated and give topological structures of interest.

When a small microsphere is inserted into the nematic liquid crystals, the molecules will align along the closed spherical surface because of the imposed alignment, as illustrated in Figure 1. Far away from the inclusion the nematic liquid crystal will be homogenous. This means that the transition region from the surface of the particle to the far field director has to be elastically deformed. Even more, it is not possible to fill this transition region with only elastically distorted director



Fig. 1. A microsphere with perpendicular surface anchoring is accompanied with a hyperbolic hedgehog defect in the nematic liquid crystal. There are actually two topological defects, which form spontaneously during the insertion of the microsphere: a virtual radial hedgehog carrying the topological charge q = +1 is formed in the center of the sphere (red dot) and a hyperbolic hedgehog defect, indicated by the blue-haze dot, is formed at a distance r_d from the surface of the sphere. This hyperbolic hedgehog carries a topological charge of q = -1, which compensates for the topological charge of the virtual radial hedgehog in the center of the microsphere. Together they form a topological dipole, also named elastic dipole.



Fig. 2. (a) A 10 μ m diameter glass microsphere with perpendicular surface anchoring in a homogeneously aligned nematic liquid crystal. Scale bar 10 μ m. (b) Schematic drawing of the director field around the microsphere. The black dot is the hyperbolic hedgehog point defect.

field. The topology of the closed surface in the inclusion requires that topological defects spontaneously appear in this transition region. One could say that the defects are topologically protected and are required due to the fundamental physical law. Indeed, one observes that these microparticles in the nematic liquid crystal are always accompanied by a topological point defect as illustrated in Figure 2.

The structure of this topological defect is of hyperbolic type and the defect is called a hyperbolic hedgehog. Similar to defects in other fields, we ascribe topological charge to defects in the nematic liquid crystals. The hyperbolic point defect is of



Fig. 3. (a) A quadrupolar colloidal particle. This is $10 \,\mu\text{m}$ silica microsphere with perpendicular surface anchoring of nematic liquid crystal molecules. Scale bar is $10 \,\mu\text{m}$. (b) Schematic drawing of the quadrupolar nematic colloid. The ring is a closed defect loop with a winding number of -1/2.

q = -1 topological charge and details of charge determination and measurements can be found elsewhere [56,57]. This topological charge of the hedgehog defect is neutralized by another topological charge located in the centre of the spherical inclusion as illustrated in Figure 1. Clearly, the structure of this virtual topological defect is radial and quite different from the hyperbolic one. Consequently, the topological charge of this virtual radial hedgehog is q = +1. This is a realization of the fundamental law of a conservation of the topological charge, as the positive unit charge of the microsphere is compensated by the opposite unit charge of the accompanying hyperbolic hedgehog.

The hedgehog and the microsphere form a topological dipole also called the elastic dipole. This is due to a large elastically distorted region of the liquid crystal around the particle, clearly visible on microscope images presented in Figure 2. Using an ordinary optical polarizing microscope, one can observe the presence of the hedgehog and also the region of elastic distortion around the spherical particle. Such observation is made possible because of the optical birefringence of liquid crystal, which makes the variation of the local optical axis (and the director) around spherical inclusion clearly visible.

In some cases, the dipolar colloidal particle transforms into a quadrupolar one, which is shown in Figure 3. This transformation occurs if the particles are confined to a very thin layer of the nematic liquid crystal. For large thickness, the particles are of the dipolar type, whereas for the very thin layer, the particles become of quadrupolar type. Quadrupolar particles are very easily distinguished from the dipolar particles. Under crossed polarisers, they give rise to an optical image with four-fold symmetry. On the other hand, the dipolar particles will give rise to an image of two-fold symmetry.

When two spherical particles of either dipolar or quadrupolar symmetry are placed close together in nematic liquid crystal one immediately observes that they interact with each other. Depending on the relative orientation and the symmetry of the particles, the force on each particle is either attractive or repulsive. In an experiment, two particles are positioned close to each other by the laser tweezers and let free to interact, as illustrated in Figure 4 for two dipolar colloidal particles. If the two dipolar particles with the same orientation of the dipoles are positioned collinearly at the separation of tens of micrometers, they start to attract each other, as shown in video snapshots in Figure 4a. Their motion can be observed in real time, and the corresponding forces between the particles can be calculated by using video tracking of their trajectories and further analysis. It turns out that the force, which is of elastic origin, is of the order of 10 pN for 1 μ m particle diameter at a separation of several micrometers. However, if the two antiparallel particles are positioned collinearly, they will repeal each other. However, if the two antiparallel particles are positioned



Fig. 4. (a) The interaction of two collinear dipolar particles with the same orientation of elastic dipoles. (b) The interaction of two collinear and antiparallel elastic dipoles.

side by side, as shown in Figure 4b, they will attract each other and form a tightly bound antiparallel pair of dipoles. The force and the corresponding elastic binding potential can be both calculated by video tracking the trajectory of the particles during their interaction. Their pair interaction energies are of the order of 1000 $k_{\rm B}T$ for micrometer diameter dipolar particles. An example of the pair binding potential is shown in Figure 5.

Whereas it is clear that the colloidal forces in nematic colloids are of elastic origin and rather easily measured, their theoretical description is not straightforward. The general theory of pair interaction forces is based on Landau–de Genns (LdG) Q-tensor approach, where one considers spatial variation of the degree of order S, as well as usual elastic deformation of the director. The numerical calculations within this general approach are quite time demanding but are necessary for the precise calculation of the structural forces. Typically, there is a maximum size limitation for the particles that are considered in the numerical calculation, which is of the order of 1 μ m. Furthermore, one elastic approximation is used. There is another approximate way to calculate the colloidal pair interaction forces, based on continuum approach, therefore neglecting spatial variation of the degree of nematic ordering. Several authors [5,11,12] used this continuum approach which is based on the electrostatic analogy and multipole expansion of the director field. The force F between the two colloidal particles, normalized to the elastic constant K of the liquid crystal can be written as a sum of three terms:

$$\frac{F}{4\pi K} = -\alpha^2 a_1^2 a_2^2 \frac{6}{R^4} + \beta^2 a_1^3 a_2^3 \frac{120}{R^6} - \alpha\beta a_1^2 a_2^2 (a_1 - a_2) \frac{24}{R^5}.$$
 (2)

Here, α and β are the dipolar and quadrupolar coefficients within the multipole ansatz, and a_1 and a_2 are the radii of the first and second particle, respectively. The centres of particles are separated by R, and we have used one elastic constant approximation with elastic constant K. The leading term in equation (2) is the attractive dipole-dipole force, which is proportional to R^{-4} . It is therefore analogous to the electrostatic force between two electric dipoles. The quadrupolar part of the force depends as R^{-6} , whereas the mixed dipole-quadrupole force scales intermediately as R^{-5} .



Fig. 5. The pair potential for two dipolar particles for parallel orientation (red circles) and antiparallel orientation (green squares). The inset shows the corresponding force plot and the best fit in the log–log scale, which is a power law with the exponent of -4.

The structural forces between particles in liquid crystals are measured using laser tweezers and the video-tracking of particles [58–73]. It was demonstrated [60], that the laser tweezing in liquid crystals is very different from optical trapping of particles in isotropic fluids. It was further demonstrated that even particles of the reflective index lower than the liquid crystal could be trapped and manipulated by focused light. The reason is orientational ordering and optical birefringence of liquid crystals which makes optical trapping dependent on the polarization of the trapping beam. In this way, it is always possible to find particular regions of liquid crystal close to the colloidal particles, where the local index of refraction is higher compared to the surroundings. Effectively, this makes it possible to trap and manipulate any kind of colloidal particle in any liquid crystal.

There are several mechanisms of optical trapping of colloidal particles in liquid crystals, which have been explored in details. This first one was just mentioned above and is based on the polarization sensitivity of the optical trapping in the birefringent liquid crystal. The second mechanism is based on the influence of the electric field of light on the local orientation of the liquid crystal. If the intensity of the laser light is high enough, the electric field of light could be so strong to reorient the molecules of the liquid crystal in the focus of the beam. This local reorientation creates a region of liquid crystal, which is different from the surrounding and we can consider it as a virtual colloidal particle. When an additional particle is brought close to this virtual colloid, they will interact through elastic forces of the distorted liquid crystal. In this way a very efficient method of manipulating colloidal particles of arbitrary material and shape is available in liquid crystals.

The third mechanism of optical trapping of colloidal particles in liquid crystals is based on local heating and even melting of the liquid crystal. A local increase of the temperature due to, for example, absorption of light in a liquid crystal, causes a decrease of the degree of order of a liquid crystal. In this way, spatial gradient of the degree of order is created, which generates a force between the heated region and the nearby colloidal particle. The reason for this force could be understood by the fact that any change in the separation between the heated region and the particle changes the elastic energy of the whole system, because the elastic constants of the liquid crystal depend strongly on temperature. This means there is a new structural force, which has its origin in the spatial dependence of the degree of orientational order. If the amount of energy supplied to the liquid crystal is high enough, it will locally melt it into a small island of the isotropic phase, surrounded by the nematic phase. This isotropic island will represent an artificial colloidal particle and the nearby nematic liquid crystal will be ordered all along the closed interface of this isotropic island. The interface between the nematic and isotropic phase will, therefore, act as a surface of the artificial colloidal particle, which will interact with the nearby colloidal particles via structural forces.

In a typical experiment of measuring the structural force between two colloidal particles, they are first brought together by the laser tweezers to a proximity of several μ m where the particles start to interact. At this moment, the laser tweezers is switched off and the particles are free to interact. During their attraction or repulsion, one takes a video recording of the whole interaction event. After that, the video recordings are analyzed by a particle tracking software, which determines the positions of both particles for each video frame with very good precision. In this way, we generate the trajectories of each of the particles in the interacting pair. From the trajectories, one can calculate the instantaneous velocity of each particle by numerical differentiation of the trajectories, which are therefore functions of time. In liquid crystal motion of particles is usually overdamped, which means that the net force on the particle is practically zero at all times. If we choose one of the interacting particles, the net force acting on this particle is:

$$F_{Stokes} + F_{structural} = 0. \tag{3}$$

In this equation, \mathbf{F}_{Stokes} is the Stokes' drag force due to the motion of the particle in the viscous liquid crystals, $\mathbf{F}_{Stokes} = 6\pi R\eta v$ for a spherical particle. This force is balanced by the structural force, generated by the second particle on the first one. By considering the above equation (3), one can clearly see that the Stokes' drag force at each time determines the unknown structural force F. This force can therefore be calculated provided we know the instantaneous velocity of the particle and the Stokes' drag coefficient. We have seen that instantaneous velocity is obtained numerically from the recorded trajectories and what is left unknown is the drag coefficient, which can be determined from an additional experiment, where the Brownian motion of the particle is recorded. By integrating this force along the trajectory of a selected particle, one can determine the pair interacting potential very precisely. Figure 5 shows an example of the measured elastic potential between two dipolar colloidal particles in the nematic liquid crystal together with their pair interacting force, presented in the inset.

It should be mentioned that forces between particles of shape different from spherical have been studied, including platelets [74–77], rods [78–80] and spirals [81,82].

3 Assembly of 2D and 3D colloidal crystals by topological defects

In nematic samples with dispersed colloidal particles, one can always observe spontaneous assembly of colloids into chains, as shown in Figure 6. Sometimes these chains spontaneously assemble into small crystals. This has led to an idea to use the structural forces in nematic colloids to assemble geometrically regular structures and crystals. These regular structures could be interesting for photonic applications, such as photonic crystals and optical cavities. Structural forces between colloidal particles in the nematic liquid crystals have therefore been studied to assemble 2D and 3D colloidal crystals of rather large size.

An example of directed assembly of a 2D colloidal crystal made of dipolar colloidal particles is illustrated in Figure 7. The laser tweezers are used to manipulate individual colloidal particles and guide their assembly into long dipolar chains, as presented



Fig. 6. Dipolar colloid spontaneously assemble into chains and small crystals.



Fig. 7. (a) Dipolar chains are obtained by sequential assembly of dipolar colloids by the laser tweezers. By continuing this operation one can assemble long dipolar chains. (b) Two chains with antiparallel orientation of their elastic dipoles are assembled into small crystals. Scale bar is $5 \,\mu$ m. (c) A colloidal crystal made of more than 270 silica micro-spheres in a liquid crystal.

in Figure 7a. In the next step, two dipolar chains with an antiparallel orientation of their dipoles are placed parallel to each other and let to assemble into a crystal, as shown in Figure 7b. By continuing this operation, one can gradually build a rather big 2D colloidal crystal, as shown in Figure 7c. One can see from the structure in Figure 7c that the crystal is composed of regularly spaced antiparallel dipolar chains. Colloidal binding along the chain is much stronger and the surface separation in this case around ~520 nm. The attractive binding potential between antiparallel dipolar chains results in ~640 nm surface to surface separation between particles in neighbouring chains. The overall symmetry of this crystal is close to hexagonal, the angle between the two neighbors is around ~61°. Dipolar colloidal crystal is extremely strongly bound together and the pair binding energy is around 1000 $k_{\rm B}T$ for silica microsphere 2.3 μ m diameter. The robustness of this crystal can be demonstrated by using laser tweezers to grab one small part of the crystal and then move the whole crystal through the liquid crystal without disrupting it.

Similar to the 2D dipolar colloidal crystals, 2D quadrupolar colloidal crystals were also assembled [83], but in this case the pair binding potential is one order of magnitude smaller and the binding energy is only several 100 $k_{\rm B}T$ compared to several 1000 $k_{\rm B}T$ for dipolar colloids. This means that quadrupolar colloidal crystals are not so robust compared to dipolar ones and can show partial and spontaneous disintegration over a longer period of time. 2D colloidal crystals of a mixture of dipolar and quadrupolar colloidal particles show an interesting wide variety of 2D motifs and good stability [84,85].

It is also possible to assemble 3D colloidal crystals out of dipolar colloidal particles and this is a very demanding task from the viewpoint of experiments [86]. Namely, one has to be able to monitor visually the colloidal structure in 3D, which can be done by using confocal optical microscope combined with optical tweezers for colloidal manipulation. Numerical calculations predicted that a 3D dipolar colloidal crystal should be assembled from individual dipolar colloidal chains which should alternate in direction for neighboring chains. This alternation of the direction of topological dipoles can be realized in the experiments for shorter colloidal chains. Typically, one can build dipolar chains up to ~ 5 dipolar particles in the nematic liquid crystal cell with perpendicular orientation at the surface. In this case, the colloidal chains shall be visible under an optical microscope as a single particle, which is followed by the neighboring particles when we change the position of the focal plane of the microscope along the depth of the cell. After building several dipolar colloidal chains, as illustrated in Figure 8b, they are led together by the laser tweezers to self-assemble into small colloidal clusters. Additional dipolar chains are added to these clusters and one obtains rather large colloidal blocks shown in Figure 8c. By taking two assembled blocks of particles, they spontaneously assemble and fuse together to form a perfect 3D colloidal crystal in Figure 8d. The structure of this crystal can be imaged by a confocal microscope and is presented in Figure 8e. As predicted, the crystal of tetragonal symmetry and shows an interesting structural response to external electric fields [86]. For example, by applying an external field, the crystal shrinks for ~ 30 percent, if the liquid crystal has positive dielectric anisotropy. If one chooses a liquid crystal with negative dielectric anisotropy, the application of the external electric field will cause the rotation of the crystal as a whole.

4 Entanglement, knotting, and linking of nematic colloids

Attractive or repulsive forces between colloidal particles in the nematic liquid crystals are caused by isolated topological defects, which belong to a particular particle and this causes aggregation and assembly of nematic colloids into different colloidal structures. In these structures, the defects are still unchanged and their number is equal to the number of colloidal particles in the assembly.

It was demonstrated in the experiment and supported by numerical calculations [87–90] that particles in the nematic liquid crystal could be bound together by a single defect loop, which "entangles" both colloidal particles, as shown in Figure 9. This is a fundamentally different structure in terms of typology because the number of typological defects has decreased from two separate loops, encircling each particle, into a single loop entangling both particles. This discovery has triggered numerous questions related to the topology and in particular the conservation of the topological charge. Namely, if at the beginning we had two separate loops, each carrying a topological charge of -1, how come a single entangled loop could carry a topological properties of loops, which has culminated in the discovery of knotted and linked defect loops [91–93].



Fig. 8. (a) Individual dipolar colloidal chains are assembled in a homeotropic nematic cell. The chains are pointing into the plain of the paper. Note the increase of the apparent brightness of a colloidal particle, which is due to the fact that this is a chain of particles. (b) Several dipolar chains spontaneously self-assemble into the small 3D block of colloidal particles. (c) Two blocks of colloidal particles spontaneously assemble into a perfect 3D dipolar colloidal crystal. (d) Larger 3D colloidal crystals also assemble spontaneously without any defect left. (e) Confocal fluorescence image of a 3D colloidal crystal shows the regular arrangement of particles in the crystal lattice.



Fig. 9. Video snapshots showing the entanglement of two colloidal particles in the nematic liquid crystal. The whole sequence of images (a–f) lasts for a couple of seconds.

It is interesting to note that the entanglement of two colloidal particles in the nematic liquid crystal has been first predicted numerically by several groups [87–89], whereas in real experiments the entanglement was demonstrated by using high-power laser tweezers [90], which could locally melt the liquid crystal into the isotropic phase, as shown in Figure 9.

In the beginning, we have two colloidal particles, each carrying a Saturn ring, which are placed close to each other as shown in Figure 9a. The laser tweezers is positioned at the sphere and the power of light is increased to the degree that locally melts the liquid crystal and we have two colloids floating in the molten island of the isotropic phase. When the light is switched off in Figure 9b, a dense tangle of topological defects is immediately created which are annihilating with time. In the end, a single loop is left in Figures 9e and 9f, which runs around colloidal particles and closes into itself. By changing the position of the focal plane, one can see that this single loop has the form of figure of the number "8", entangling both colloidal particles. The two particles are very strongly bound together. If one wants to increase the separation between the particles, one has to increase the length of the defect loop, which requires additional energy. This entanglement results in pair binding energies of $10^4 k_{\rm B}T$ for several μ m-diameter colloidal particles. The forces due to entanglement are also quite strong and of the order of ~30 pN.

It is interesting that only the entanglement of linear colloidal structures could be obtained in a non-chiral nematic liquid crystal and no entanglement of 2D colloidal structures was observed in a plane. However, when one is using chiral nematic liquid crystal instead of non-chiral one, interesting new phenomena are observed, which are related to the spontaneous entanglement of colloidal particles in the plane of the cell. Some examples of the spontaneously entangled structure of several colloidal particles are shown in Figure 10. The structures were obtained in a liquid crystal cell, where the liquid crystal was chiral and twisted for 90° when going from one surface of the cell to the other.

By careful observation of the structure of the defect loops running around the colloidal particles, one can distinguish that the colloidal pair in Figure 10a is actually a "figure of 8" loop, which is slightly twisted due to the twisted environment. The trimer in Figure 10b is also entangled by a single loop, which is running around



Fig. 10. Colloids in 90° twisted nematic cell show spontaneous entanglement into (a) dimers, (b) trimers and more complicated linked and knotted structures (c-f).

the colloidal particles, twisting in between and closing itself into a single loop. In terms of topology, the dimer and the trimer in Figures 10a and 10b are the unknots. However, interesting situation is observed in Figure 10f, where 7 colloidal particles are entangled by a single or perhaps several loops. Similar interesting topology is observed for a larger number of colloidal particles, presented in Figures 10c-10e. One is asking whether the loops in these figures are actually knotted and linked defect loops? In order to answer this question, one performs the Reidemeister moves [94], which are illustrated in Figure 11. After taking a photo of the colloidal structure, one is drawing the structure of the defect loop(s) entangling the colloidal cluster as shown in Figures 11a and 11b. After that, the Reidemeister moves are performed and shown in Figures 11c-11f, which means that the loop can be made smoother and untwisted, but without cutting and rejoining it. The result for the colloidal object in Figure 11a is the Trefoil knot [94]. This means that there is a single defect loop running around colloidal particles in Figure 11a, which is actually of the form of the Trefoil knot. This is a single defect loop with three crossings. A similar analysis for the colloidal cluster in Figure 12a shows that this object is a Hopf link. Instead of a single loop, two separate loops are entangling the cluster of six colloidal particles, as shown in Figure 12b. These two loops are not isolated, but are linked together.

One can systematically analyze the appearance of knots and links on a 2D array of particles, organized in p rows and q columns, as illustrated in Figure 13. There is clearly an interesting pattern in the topological properties of defect loops entangling regularly placed colloidal arrays. The resulting structure is consequently alternating between knots and links and they are all Torus knots and links as they could be formed on the surface of the Torus. We should mention here that the first report on knotted and linked defects was presented by Y. Bouligand in 1970s [95–97]. His careful observations of cholesteric liquid crystalline textures under an optical microscope were one of the first topological studies of liquid crystals and reported some of the phenomena, which were rediscovered more than 30 years later by using new experimental techniques such as the laser tweezers and the fluorescent confocal microscope.

The beauty of topological structure of defect lines entangling colloidal clusters in two dimensions can be systematically investigated by using the laser tweezers, which



Fig. 11. (a) An example of 7 colloidal particles, entangled by a single defect loop. After extracting the shape of the loop (b), Reidemeister moves are performed (c-f), which show that the loop is actually the Trefoil knot.



Fig. 12. An example of the six colloidal particles entanglement by two linked defect loops, forming a Hopf link.

made it possible to cut and rejoin defect loops on the colloidal array, as presented in Figure 14. Here the laser tweezers' optical power is increased to the level that the laser light locally influences the liquid crystal structure and causes the topological defect lines to cut, insert and rejoin. This is a kind of microsurgical operation, performed under an optical microscope and enables one to obtain and analyze a huge variety of topological structures obtained in a chiral colloidal system.

A colloidal array of 4×4 silica particles was used to systematically analyze using the computer software all possible topological structures that could be generated [91]. It turns out that there is a nearly equal amount of possible knots and two component links, and there is a minority of three and four component linked structures. The more complex structures are less probable. For example, one finds a nearly 10 percent probability for the formation of the Trefoil knot whereas the probability for the formation of a knot with 8 crossings is less than 0.1 percent. Interestingly, there is an exponential decrease of the probability for the formation of the knot with an increasingly large number of crossings. Similar dependence is observed for the two component links [91]. Special attention has been paid to the structure of defect



Fig. 13. Examples of Torus knots and links obtained on a $p \times q$ array of colloidal particles in a twisted nematic liquid crystal. (a) 3×3 particle array with the extracted knot projection. After Reidemeister moves are performed, one obtains a knot diagram which shows that these nine particles are knotted by a single defect loop which is a Trefoil knot. By adding additional three particles which give us a 3×4 particle array, one obtains two interlinked defect loops which represent a Solomon link. (b) By systematically adding additional rows of particles, one obtains a Pentafoil knot and for additional three particles forming 3×6 particle array a Star of David is obtained.



Fig. 14. Laser tweezers is used to cut and rejoin or even insert additional colloidal particles into the existing colloidal array. By cutting and rewiring a particular crossing of defect lines in the colloidal array, the global topology of defect loops is changed at will.

loops, which are entangling the colloidal particles, because they determine the topological charge of this assembly. Topological properties and especially the role of the self-linking number of these loops were extensively studied by Čopar et al. [98] in the context of nematic braids.

It is interesting that in many fields across physics [3,56], chemistry [99], and even biology [100], topology is becoming an important aspect of material properties in the last decade. For example, knotting of fields has been studied and observed in optics [101,102], where the electromagnetic waves were found to be knotted and linked. Tying knots in the light fields have been analyzed theoretically and isolating optical vortex knots were constructed from knotted complex wave fields. Knotted vortices and pairs of vortex links have been generated and analyzed in hydrodynamic experiments in water [103,104]. Very strong interest has been observed recently in topological properties of light for application in topological photonics [105]. Here, Möbius strips of optical polarisation have been demonstrated [106], and the topological insulator lasers were constructed [107]. These new devices based on topology find interesting application across different fields of science. In quantum optics and cold atoms tying on quantum, knots were demonstrated [108], which is one of the first demonstrations of knotting the quantum matter, which is, in this case, the order parameter of a spinor Bose-Einstein condensate.

In liquid crystals, studies of topological properties have been particularly rich in the past 10 years. Especially interesting are chiral systems, which by their chiral nature promotes complex topology of the order parameter field. In this respect, there is a strong analogy between the topology of the chiral nematic liquid crystal and chiral magnetism. In some cases, there is nearly one-to-one correspondence of the topological states, which have been observed. This is particularly true for Skyrmion and related structures. In the liquid crystal, Skyrmions are localized swirling structures, which are smoothly embedded in the far field of the homogeneous liquid crystal. Skyrmions and torons were first directly observed in an experiment by Smalyukh et al. [109] in chiral nematic liquid crystal confined to a rather narrow gap with perpendicular surface anchoring of liquid crystal molecules. Due to the tight confinement, the liquid crystal has to be unwound into a homogeneous texture, which is energetically frustrated. This frustration results in a spontaneous formation of localized twisted structures, appearing as small bubbles of a liquid crystal under an optical microscope. These bubble-like structures were observed already in the 1970s but were not analyzed in detail, because there were no experimental tools available for measuring and reconstructing local director structure in such a deformed object. Smalyukh et al. [110–112] have used a Fluorescent Confocal Polarisation Microscope (FCPM) to analyse the internal structure of torons. Using FCPM, one can locally measure and determine the orientation of fluorescent molecules, which are dissolved into the liquid crystal. They locally align with the nematic liquid crystal director and serve as optically polarized antennas, which can be seen and detected by FCPM. Using this method, it was possible to demonstrate many different topological structures (also called excitations), such as Skyrmions in thin layers of a chiral nematic liquid crystal, Torus and Hopf fibrations [113] in thin layers of chiral nematic liquid crystal. FCPM has been used to analyze the director structure in chiral nematic droplets where the numerical calculation predicted the existence of knotted and linked defect loops. Instead of knots and links, Posnjak et al. [114] demonstrated the existence of point defects of unit charge, organized in the interior of the droplets as strings of unit charge point defect. In highly twisted materials, they observed point defects of higher than unit topological charge, such as three and five unit charges [115].



Fig. 15. Examples of polymer particles produced by 2-photon polymerization: (a) polymer coils, (b) Koch star particle.

5 Topological colloids

Studies of liquid crystal colloids have been mostly concentrated on colloidal particles of spherical or rod-like shape. The reason for this was ease of their fabrication; for example, spherical microparticles can be fabricated with great precision from glass or polymeric materials. Rod-like particles can be fabricated by cutting fibers or by a directed growth of elongated objects. In terms of topology, micro-rods are of the same shape as micro-spheres, because they have no "hole". In topology, the number of "holes" is measured by the genus g of the particle. For example, a sphere has the genus q = 0, and the torus has the genus q = 1. During the last ten years, new 3D additive manufacturing techniques have been developed, based on precise photopolymerization, controlled at nanometer resolution in 3D. Although this restricts the available materials to photosensitive polymers, it also open possibilities of manufacturing micro-objects of arbitrary shape. For example, particles in the shape of coils and spirals have been fabricated and studied in liquid crystals [82]. They both have genus q = 0 and are topologically of the same shape as spheres and rods, but their helical shape is responsible for a novel type of pair interaction of liquid crystals. Recently, particles of the shape of knots and links have been made by photopolarization and inserted into liquid crystals [116]. As the liquid crystal has to conform to the shape of polymer knots and links, it forms novel topological liquid crystal structures of defect loops, which have been identified as knots and links of topological defects. Handlebody polymeric particles have been fabricated both by photolithography of thin silica and photo-polymerization [117]. These particles have genus g depending on the number of "handles" and bring the new topological structure of point defect and loops that have to obey the law of conservation of topological charge. Koch star microparticles have been fabricated from polymers and inserted into the liquid crystal [118]. These particles are of the same form as torus particles and have genus q = 1. This means that the topological charge of Koch star particles should be equal to that of the Torus particles. However, Koch star particles do not have a smooth closed surface. Instead, the surface shows discontinuities at kinks where the surface breaks and forms a star. Some examples of complex shaped colloidal particles made by photopolymerization using intense laser beam are shown in Figure 15. The kinked surface of the Koch star particle induces the formation of topological defects at points where the surface tangent is discontinuous. Accordingly, the number of possible topological defects on Koch star particles is very large, however, the sum of all charges of these topological defects should sum-up to the value equal for smooth Tori.



Fig. 16. (a) An example of a saddle-like surface, and the planes of principal curvatures to calculate the local Gaussian curvature at each point of the surface (Wikimedia Commons contributors, Wikimedia Commons, the free media repository, 2018). (b) A Torus has negative Gaussian curvature on the "inner" parts of the surface and a positive Gaussian curvature on the "outside" area (reproduced from Wikimedia Commons, the free media repository, 2018.)



Fig. 17. A sphere has a genus g = 0. By adding one handle to the sphere, we obtain an object that is of the same form as a coffee mug. This coffee mug can be smoothly transformed into a Torus. Note that smooth transformations of surfaces are allowed in topology.

The importance of the genus g colloidal particle was first stressed by Senyuk et al. in an article entitled "Topological Colloids" [117]. The authors made silica handlebodies from thin slices of silica and the number of holes of handlebodies was systemically varied. Using the FCPM, they observed a systematic pattern in the number and shape of topological defects, carried by these handlebodies. In turns out that the topological charge of handlebodies is directly related to the total Gaussian curvature of the handlebodies and therefore also to the genus g of these particles.

Let us imagine a saddle like surface illustrated in Figure 16a. We choose a point on the surface and construct a vector normal to the surface at that point. Furthermore, we construct mutually orthogonal planes, intersecting at the chosen point and define the local Gaussian curvature K, which is a measure of the local curvature of the surface:

$$K = \frac{1}{R_1} \cdot \frac{1}{R_1}.\tag{4}$$

In this equation, R_1 and R_2 are the local principal radii at the selected point. For a sphere, local Gaussian curvature is positive and equal to $1/R_2$, which is positive, everywhere. For a cylinder, local Gaussian curvature is K = 0 everywhere on the surface. For a rotational hyperboloid, the local Gaussian curvature is negative and equal to $1/(R_1 + R_2) < 0$ everywhere on the surface. In contrast to a sphere, a cylinder and a hyperboloid, which have the constant local Gaussian curvature at each point of the surface, a torus is an example of a surface that has a negative curvature in some places and a positive Gaussian curvature in another parts, as illustrated in Figure 16b.

Next, we define the Euler characteristic χ of a surface, which is just the integral of the local Gaussian curvature over the surface S:

$$\chi_S = \frac{1}{2\pi} \cdot \oint_S K \cdot dS. \tag{5}$$

On the other hand, we can calculate the total Gaussian curvature by performing the surface integral of the unit vector $\boldsymbol{\nu}$, which is just a local normal to the surface.

$$\chi_S = \frac{1}{2\pi} \cdot \oint_S d\theta d\varphi \cdot \nu \left[\frac{\partial \nu}{\partial \theta} \times \frac{\partial \nu}{\partial \theta} \right]. \tag{6}$$

The Euler characteristic of the surface is related to the Genus g of the surface via the Gauss–Bonnet theorem:

$$\chi = 2(1-g). \tag{7}$$

Here the genus g is actually the number of handles of the particular handlebody. For example, the sphere has no handles and the g = 0. If we attach one handle to a sphere, as shown in Figure 17, we obtain a coffee mug after some smooth modification (i.e. morphing). This coffee mug can be smoothly transformed into a Torus, which is an object with one hole and genus g = 1.

Let us now consider a simple handlebody in a form of a sphere or a Torus, embedded into a vector field and this field is forced to be perpendicular to the surface of the inclusion everywhere, as shown in Figure 18. Let us remember that the Euler characteristics is equal to $\chi = 2$ for a sphere and is equal to $\chi = 0$ for a Torus. We now discuss the connection between the Euler characteristic of a given closed surface defined and the topological charge of a vector field \boldsymbol{n} . The topological charge of a vector field, which is forced to be perpendicular at each point of a closed surface, is measured by the method of Gauss, i.e. by performing the surface integral of the flux of the field:

$$m_c = \frac{1}{4\pi} \cdot \oint_S d\theta d\varphi \cdot n \left[\frac{\partial n}{\partial \theta} \times \frac{\partial n}{\partial \theta} \right].$$
(8)

One can immediately observe that the definition of topological charge m_c of a vector field on a closed surface is directly related to the Euler characteristics of that closed surface and is, therefore, also related to the genus g of the surface:

$$m_c = \pm \frac{\chi}{2} = \pm (1 - g).$$
 (9)

Let us now interpret the relation of the topological charge of the surface and the topological charge of all defect of the nematic director field surrounding the surface of the inclusion. We start from the defect-free nematic liquid crystal with zero topological charge. Now, we insert an object into the liquid crystal that is characterized by its closed surface. Because the inclusion forces the liquid crystal molecules to align, let us say perpendicular at each point of the surface of the inclusion, the surface adopts a topological charge as defined by equation (9). Here, the local director n on the surface has the same role as the local normal to the surface. This means that this surface field has now a topological charge must be preserved at all times. Accordingly,



Fig. 18. (a) A vector field is forced to align perpendicular to the surface of a sphere. If you consider all possible orientations of the vector as you are visiting all points on the surface, you see that it points to all possible directions of a 3D space only once. (b) A vector field is forced to align perpendicular on the surface of torus everywhere. If we are following all possible orientations of this vector as we visit all points on the surface of torus, you see that each possible orientation in 3D space is met twice, once at the outer surface and once at the inner surface.

the nematic liquid crystal surrounding the included object must develop topological defects, which are carriers of the topological charge. The total charge m_d of all topological defects surrounding the inclusion has to be, for topological reasons, opposite to the topological charge of the included surface:

$$m_c + m_d = 0 \tag{10}$$

and

$$m_d = \mp \frac{\chi}{2} = \mp (1 - g). \tag{11}$$

We therefore, get a simple answer to what should be the topological charge of all defects accompanying the closed surface of colloidal inclusion. For example, the topological charge of a sphere is equal to +1. Because the topological charge of the sphere, inserted into the liquid crystal, has to be compensated to zero, a hyperbolic topological defect has to be created in the vicinity of the sphere, compensating the +1 charge of the sphere to zero. This gives us the answer why a micro-sphere with a perpendicular surface anchoring of liquid crystal molecules is always accompanied by a point hyperbolic hedgehog defect. Therefore, the defect is topologically protected and simply has to be there as long as the sphere is there.

An interesting situation is observed when considering the total topological charge of defects accompanying toroidal particles, which is inserted into the liquid crystal and induces perpendicular surface anchoring of liquid crystal molecules. Because of the zero topological charge of the Torus $m_c = 0$, the total charge of surrounding topological defects must be zero. Now, there must be some topological defects surrounding the inserted Toroidal particles simply because it is not possible to transit smoothly from the surface of the Torus to the far field homogeneous director. Clearly, topological charges must be created, but their topological charge should be mutually compensated. This means that the inserted Torus shall be accompanied by at least two topological defects with the opposite topological charge. This was indeed observed by studying the number and the structure of topological defect for Toroidal particles using the FCPM [117]. It was observed that the Toroidal particle is indeed always accompanied by a pair of oppositely charged topological defects, which are



Fig. 19. Millions of liquid crystal droplets can be made in a fraction of a second by simply mixing liquid crystal and an immiscible fluid. The droplets are viewed under crossed polarizers and the colours appear due to optical birefringence and spherical shape of the liquid crystal.

both hyperbolic hedgehogs. This was generalized to handlebody colloids with higher genus up to g = 5 which was a direct experimental proof of the Gauss-Bonnet theorem in liquid crystals. The work of Senyuk et al. [117] illuminates very nicely the problem of assigning the topological charge to hedgehogs in nematic liquid crystals. It demonstrates that the same structure of hyperbolic point defect can have two different signs of the hedgehog charge, and the sign depends on the overall nematic texture. This problem of the sign of the topological charge of hedgehogs in nematic liquid crystals was discussed in detail by Alexander et al. [56].

6 Light and topology: microdroplets, microlasers and fibres made of liquid crystals

During the last ten years, there has been a considerable progress in the new subfield of the soft matter related to photonics of soft and living matter. In liquid crystals, new ideas emerged around the year 2009, when a small liquid crystal device with a function of an optical microresonator was proposed and demonstrated [119]. This device is actually just a small droplet of a liquid crystal made by dispersing liquid crystal material into an immiscible carrier fluid, which is usually water, polymer, etc. When such an emulsion is formed, millions of small droplets of liquid crystal are formed and they are perfectly spherical with perfectly sharp and well-defined interface between the interior liquid crystal and the exterior fluid. An example of such an emulsion is shown in Figure 19, showing droplets of a chiral nematic liquid crystal in water.

In the past, the emulsion of liquid crystal has been intensively studied in the context of polymer dispersed liquid crystal (PDLC), where submicrometre droplets are dispersed in a polymer. The index miss-match causes such material to appear milky due to strong scattering of light on submicrometer droplets. An electric field changes the refractive index of a droplet, induces better optical matching and renders



Fig. 20. (a) A radial droplet is obtained when the surface anchoring of liquid crystal molecules is perpendicular. (b) A bipolar droplet of a nematic liquid crystal with parallel surface anchoring.

the material's transparency. It should be noted that typical droplets in a PDLC material are at least 1 to 2 orders of magnitude smaller than the droplets considering for applications as photonic microdevices. A typical diameter of a droplet we are considering in the continuation is larger than 10 μ m.

It turns out that droplets of thermotropic liquid crystals dispersed in another immiscible fluid, such as water, are always perfectly spherical in spite of the elasticity and elastic deformation of a liquid crystal material inside the droplet. The surface energy of a typical droplet of a nematic liquid crystal droplet in water $F_s = 4\pi R^2 \sigma$ is several orders of magnitude larger compared to the typical elastic energy $F_{elast} \approx K \cdot R$, which is due to elastic distortion of the liquid crystal inside the droplet. Liquid crystals that are based on water dispersions of molecules or nanorods or rod-like viruses, where the surface tension of the surrounding water is usually very low. In this case, the elasticity of the liquid crystal of the interior becomes of significant importance. Instead of the spherical shape of thermotropic liquid crystal droplets, one obtains a "tactoidal shape" of water-based liquid crystal droplets in water. The internal structure of nematic droplets depends on the surface anchoring at the interface to the external fluid and are typically classified into radial droplets and bipolar droplets as shown in Figure 20.

A radial nematic droplet is obtained when the anchoring of a liquid crystal at the interface is perpendicular. Because of the spherical confinement, there must be a single topological defect of a radial structure carrying a topological structure +1 in the centre of the droplet. A bipolar nematic droplet is obtained for parallel surface anchoring of liquid crystal molecules at the interface. In this case, the topological charge +1 is distributed between the two surface boojums residing at the interface. Interestingly, these dipolar droplets are actually very similar to what is obtained in the process of cell-division. The spindle fibers are organized in a similar way as the nematic director in dipolar droplets and they emanate from the two spindle poles. These poles, therefore, remind us of topological defects in dipolar liquid crystal droplets. The internal structure of a nematic droplet will depend on the diameter of the droplets, as shown in Figure 20b.

Clearly, there is a competition between the elastic energy of the interior of the nematic droplets and the surface anchoring energy. If the diameter of the droplet is large, the total elastic energy will be small compared to the surface energy, because it depends linearly on the diameter of the droplet. In this case, a nematic droplet shall be uniformly split with a direct singular in the centre which is a radial hedgehog defect. If the droplet's diameter is decreased, the surface energy will decrease proportionally to R^2 , whereas the elastic energy will decrease linearly in R. There will clearly be a



Fig. 21. Examples of a nematic droplets with perpendicular surface anchoring. For large droplets, the topological defect will appear in the centre of a droplet. In smaller droplets, the interior will be practically homogeneous and the surface anchoring will break.

crossover size where the surface anchoring requiring perpendicular surface anchoring will be too small compared to the elastic energy. In other words, the elastic stiffness of the confined liquid crystal will be too high and the result will be breaking of the surface anchoring. These intermittent droplets, shown in Figure 21, shall be slightly inhomogenous in their interior. There will be no topological defect as the energy cost for their creation is too high compared to the energy required for the breaking of the surface anchoring. Finally, very small droplets of a nematic liquid crystal shall be practically homogeneous in their interior and their surface anchoring will play no role. There is a crossover size for the transition between large droplets with the topological defect of small droplets with the nearly homogenous interior size, which is equal to the surface extrapolation length defined in equation (1). Typical extrapolation lengths are of the order of of 100 nm. It is clear that droplets larger than 1 micron could be very rich in their topology, since at least one topological defect has to be present due to confinement of a liquid crystal by a closed surface. It turns out that the topology of larger droplets made of chiral nematic liquid crystals are actually very rich in topology. On the other hand, very small droplets that are usually used in PDLC materials are obviously poor in their topology and their only function is to respond to external field and change the external property of the dispersion.

A great deal of interest has been shown in studying optical properties of nematic droplets of a typical diameter of 10 micrometers, which are freely floating in a carrier fluid. If one chooses a carrier fluid with a refractive index, which is much smaller than both refractive indices of the nematic liquid crystal and the crystal is aligned perpendicular to the interface, one obtains a radial nematic droplet, which is of perfect optical quality in the interior. Moreover, the interface between the liquid crystal and the external fluid can be made very sharp and optically completely flat, because it is stretched by the surface tension. Such a radial birefringent microsphere is actually a perfect optical micro-cavity that is able to confine the light in its interior.

Optical microcavities are at the focus of current researching photonics because they are basic optical microelements with the function to confine light. There are two basic mechanisms of confining the light in an optical microcavity. The first mechanism uses a total internal reflection between the high index interior of the droplet and the low index of the surrounding matter. Imagine that light is created inside the microcavity and it propagates towards the interface. If the angle of incidence at the point of reflection is high enough, all light will be totally reflected from the interface and shall bounce-off to the interior. This process will repeat by subsequent total internal reflections. If this light reaches the place of the first reflection with the same phase, we shall create a standing wave along the circumference of the droplet. These waves are known as Whispering Gallery Modes (WGMs) and were first studied as acoustic resonance modes in shells and domes of cathedrals. In photonics, WGM resonators are usually made from solid materials in very different forms such as spheres, disks, tori and race tracks.

The second mechanism for light confinement uses an optically periodic structure with a photonic band gap serving as a perfect Bragg mirror due to the interference of waves. A typical Bragg microcavity will be made of an optically transparent material, which is surrounded by a periodic structure. If waves are created in the interior of the microcavity, this light will be reflected from the surrounding Bragg structure simply because it cannot propagate in this optically forbidden region. Bragg microcavities are usually made from thin slices of a high refractive index material, and the Bragg reflector structure is made by producing holes organized in a prescribed geometric pattern. The holes are filled with a lower reflective index material, such as air or even left empty, and this periodic reflective structure of a high/low reflective index will reflect light within a certain frequency band.

Whereas the solid microcavities made by photolithography of solid material have the advantages of being currently technologically mainstream, there are obvious advantages of photonic elements made of soft matter. Firstly, they have perfectly flat optically interface which can be easily tuned due to softness and fluidity of the elements. Secondly, soft matter elements, such as microcavities, are very easy to produce due to their fluidity and the capacity to self-organize. For example, the internal structure of nematic microdroplets will be self-organized practically instantaneously at the moment when the droplet is formed. Thirdly, soft matter materials can be easily tailored by mixing, doping and labeling with fluorescent dyes. All these steps are usually performed at room temperature for soft matters, whereas solid-state fabrication requires higher temperatures and many technological steps.

Let us now illustrate the advantages of soft matter photonic elements on a very simple example: A tunable optical microcavity is made of a radial nematic droplet dispersed in water. Its diameter is typically 10 micrometers. An example of such droplet is shown in Figure 22a together with the schematics of molecular distribution in the droplet's interior as illustrated in Figure 22c. A small amount of fluorescent dye is added to the liquid crystal and the radiative dipole of dye spontaneously aligns along the local nematic director.

When the edge of the radial nematic droplet is illuminated with the argon laser, it induces the fluorescence of fluorescent dye inside the droplet. One can clearly see a rather strong fluorescent light, which is radiated from the other side of the droplet. Clearly, the fluorescent light is circulating inside the droplet in the form of WGMs. If one analyses the spectrum of light emitted from the illuminated radiated nematic droplet, one can clearly see the presence of sharp spectral lines on the broad fluorescence background as shown in Figure 23. The WGM resonances are very narrow with a typical half-width of 0.05 nm, which in terms of frequency corresponds to 15 GHz width. The corresponding quality factors of these nematic microcavities is quite high and is above $Q > 10\,000$. It turns out that the WGM resonances are identified as transverse magnetic modes and the mode numbers can be described using a simple optical model.

Since liquid crystal responds strongly to the external fields, we expect that the applied electric field will change the director structure of the droplet's interior. Accordingly to this field-induced change, one expects a corresponding change of the optical path of the WGMs, which should change their frequency. Indeed, this is very



Fig. 22. (a) A microscope image of a nematic droplet in water. (b) The same droplet under crossed polarizers. (c) The schematic drawing of distribution of liquid crystal molecules. (d) Fluorescent imagine of the same droplet as in (a) when the edge of the droplet is illuminated with the green line of the argon laser (black cross). (e) An artist's impression of the excitation of WGMs in a radial nematic droplet (orange sections) by the green laser light.



Fig. 23. WGM resonances in a radial nematic droplet. (a) A single set of WGM resonances is observed in a 10.1 μ m droplet of E12 in PDMS, corresponding to the lowest radial modes (n = 1) with TM polarization. (b) In a larger radial birefringent droplet (12.6 μ m) second radial modes appear with n = 2. The inset shows details of a WGM spectral line in a 53 μ m diameter E12 droplet. The linewidth is approximately 0.055 nm, and the LC cavity *Q*-factor is of the order of ~12.000. This spectra was measured using a high-resolution micro-Raman spectrophotometer.

clearly observed in the experiments, where an individual radial nematic droplet is subject to an external electric field of a typical strength of 10 volts applied across 10 micrometres, as shown in Figure 24. Under the application of the external electric field, the radial hedgehog, which is residing in the centre of the droplet, splits into a small ring. By increasing the electric field, it is pushed towards the surface of the droplet. At a very high field, the interior of the droplet is practically homogenous with a surface defect ring, which is pushed towards the interface by the elasticity. Considering the circulation of WGMs inside the droplet, there is the change of the optical path for the particular mode. The change appears because there is the change of orientation of liquid crystal molecules near the surface ring, where the local dielectric constant effectively decreases. This makes a change in the resonance condition, where the wavelength of the mode decreases due to decreasing optical path. The blue-shift induced by the external electric field is quite high and is of the order of 5 nm for a field



Fig. 24. The application of an electric field changes the interior structure of the radial nematic droplet and induces a frequency shift of the WGMs. (a) A nematic droplet at zero electric field between crossed polarizers. (b) The electric field is turned-on, which causes the reorientation of the liquid crystal. (c) At low fields, the region near the interface of the droplet is still aligned perpendicular to the surface. (d) At higher fields, the topological defect is expelled to the interface where it forms a surface ring. Shaded regions indicate a local change in the molecular orientation, pointing to a local change of the reflective index and optical path.

strength of 1 volt per micrometre, as shown in Figure 25. The electric field tuneability of liquid crystal microcavities is nearly two orders of magnitude larger compared to similar effects in solids, highlighting clear advantages of photonic microelements made of soft matter; in this particular case of the nematic liquid crystal.

There are many more examples of liquid crystal photonic microelements, which have been discovered and studied for the past ten years, including tunable microlasers, sensors made of nematic microcavities, and fluid microfibers that spontaneously assemble and grow [120–122]. Many of these elements can be permanently fixed by photopolymerization preserving their photonic functionalities. One of the most unusual example of a soft matter photonic device is a 3D microlaser [122]. This is a droplet of a chiral nematic liquid crystal of typically 10 micrometre in diameter, which emits laser light in all direction when it is remotely excited by another laser. One could foresee many different applications of soft matter micro-photonic devices in the near future. In particular, there is a strong new line of research of soft matter micro-photonics which is directed towards the biophotonics [123].

7 Conclusion

The field of liquid crystals has witnessed a revival in the last 15 years, where many new ideas, fundamental properties, and phenomena were discovered, promising interesting application in future devices. Liquid crystal colloids and dispersions have opened a new line of research in experimental topology, where the structure and dynamics of defects could be studied in real space and in real time. Many fundamental theorems of topology have been proved in direct experiments, for example, the conservation of



Fig. 25. The electric field dependence of the wavelengths of the WGMs in a 16.1 μ m diameter microdroplet of an E12 nematic liquid crystal and SPP-106 fluorescent dye. Color scale indicates intensity of detected light. Note the enhancement of the resonances at discrete wavelengths.

the topological charge and Gauss-Bonnet theorem for defects accompanying handlebodies in liquid crystals. This research could not be possible without the invention of new experimental methods, in this case, fluorescent confocal polarized microscopy (FCPM), two-photon polymerization and the laser tweezers. By using the FCMP, it was for the first time possible to reconstruct the director field in complex geometries, such as chiral nematic droplets or handlebodies immersed in a liquid crystal. By using laser tweezers, it was for the first time possible to manipulate and move individual colloidal particles and study colloidal forces and assembly. By using two-photon polymerization, it was for the first time possible to manufacture micrometre-sized objects of complex geometry and topology, which were then immersed in the liquid crystals and their topological properties and pair interaction forces were studied.

In addition to fundamental aspects, the field of liquid crystal colloids and dispersions has demonstrated many new directions towards applications. This includes tunable optical microcavities and lasers, microsensors based on interfacial phenomena and liquid crystal dispersions with magnetic and novel optical properties. Dispersions of nanoparticles in the nematic liquid crystals have resulted in spontaneous magnetization of a nematic liquid crystal and quite recently the biaxial nematic liquid crystal made of two interconnected "sublattices" of liquid crystal order and order of nanorods have been discovered. There are many other examples where the liquid crystal colloidal physics opens new and exciting pathways towards new phenomena which unfortunately cannot be discussed here.

This mini-review is based on lectures delivered at Geilo School 2017 Physics Inspired by Living Matter: Dynamics, Topology and Functionality and is a tutorial by nature. Due to length limitations, this work cannot capture and include many important research topics and references, but can only give a limited overview of this exciting and still rapidly developing subfield of liquid crystals. For this reason, many references and important publications could not be included.

The author wishes to acknowledge contribution of Miha Škarabot, Slobodan Žumer, Miha Ravnik, Andriy Nych, Ulyana Ognysta, Simon Čopar, Matjaž Humar and many others with whom the author collaborated during the last 15 years in this field. The support of Slovenian Research Agency (ARRS) through contracts P1-0099 and J1-6723 is acknowledged.

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