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TOPICAL REVIEW

Direct and inverted nematic dispersions for soft matter photonics

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Abstract

General properties and recent developments in the field of nematic colloids and emulsions are discussed. The origin and nature of pair colloidal interactions in the nematic colloids are explained and an overview of the stable colloidal 2D crystalline structures and superstructures discovered so far is given. The nature and role of topological defects in the nematic colloids is discussed, with an emphasis on recently discovered entangled colloidal structures. Applications of inverted nematic emulsions and binding force mechanisms in nematic colloids for soft matter photonic devices are discussed.

(Some figures in this article are in colour only in the electronic version)

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1. Introduction

Nematic colloids are dispersions of microparticles in a nematic liquid crystal [1]. Interestingly, nematic colloids were considered a long time ago in the pioneering studies of the alignment of nematic liquid crystal molecules at a liquid crystal (LC)–air interface [2]. Small particles were used to ‘decorate’ the LC–air interface and it was observed that the microparticles

spontaneously organized themselves in chain-like structures, which were following the local direction of the nematic liquid crystal molecules at the interface. The method of decorating the LC–air interface was therefore useful for visualizing the nematic liquid crystal ordering pattern. At that time, similar decoration methods were used to visualize the lines of the spontaneous polarization and magnetization in solids. The seminal study of Poulin *et al* on nematic emulsions made in 1997 [3] has attracted substantial interest. They were studying the behavior of microdroplets of water, which were dispersed by mixing a small amount of water in the nematic liquid crystal. They observed that, quite interestingly, the water microdroplets spontaneously organized themselves into well defined chain-like structures, which were following the direction of the alignment of liquid crystalline molecules. It was clear from this experiment that the nematic liquid crystal generates forces between immersed microparticles, but the experiment triggered many fundamental questions, such as that of the role and nature of topological defects [4, 5], which were clearly responsible for the stability of chains of water droplets and that of why these droplets did not coalesce.

The motivation for recent studies in the field of nematic colloids arose from curiosity about whether colloidal particles in the nematic LC could organize themselves in 2D and

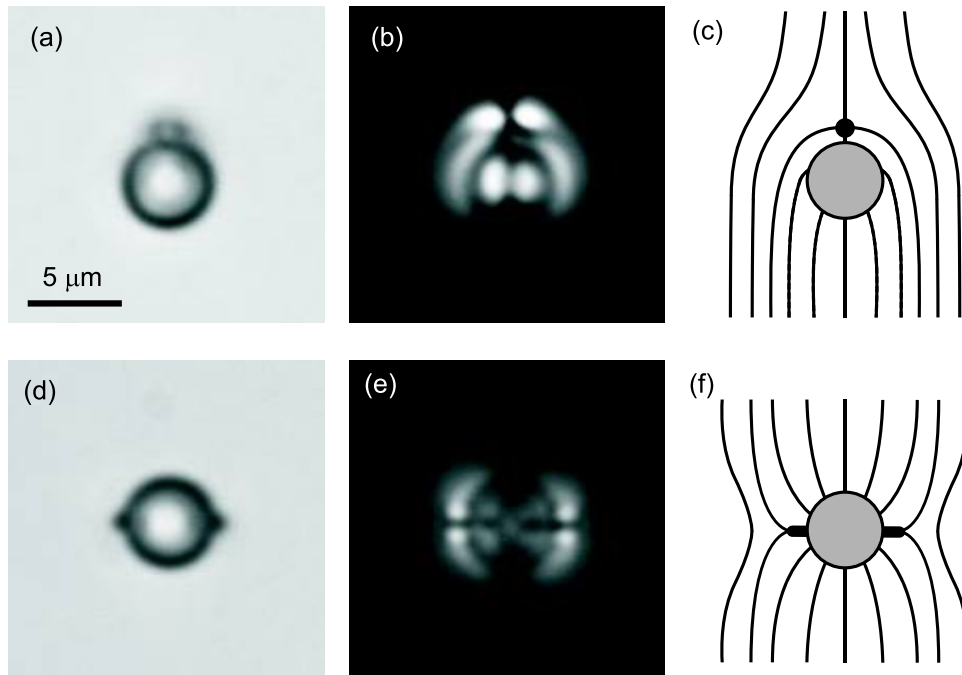


Figure 1. Nematic colloids under an optical microscope. (a) A $4.7\ \mu\text{m}$ dipolar nematic colloidal particle, whose surface orients the nematic LC perpendicularly. (b) The same particle under crossed polarizers. The nematic LC is distorted even far away from the particle. (c) A schematic representation of the lines of orientation of the nematic LC. (e) A quadrupolar nematic colloidal particle with its Saturn ring lying perpendicular to the plane of the image. (f) The same particle as in (e), but now under crossed polarizers. (g) A schematic representation of the nematic LC ordering around a topological quadrupole.

possibly 3D, which could make them interesting for photonics applications, such as obtaining photonic crystals [6]. These materials, which are characterized by the spatially periodic arrangement of dielectric objects, are the subject of intensive research with the aim of finding a way to achieve assembly or self-assembly mechanisms for photonic devices, which could control the spatial flow of photons. On the fundamental side, the motivation came from the unusual nature of topological defects in nematic liquid crystals and the general topological properties of the ordering field in nematics [5].

In this paper, we review recent developments in nematic colloids with an emphasis on structural forces, which are responsible for a fascinating variety of 2D colloidal crystals and superstructures observed in this system. We propose that structural forces in nematic colloids could provide a route to a novel class of photonic devices, based on soft matter microstructures. This review is mainly focused on work conducted in our laboratory, but an extensive list of references is given, detailing the achievements of other groups as well.

2. Structural forces and colloidal ordering in nematic colloids

2.1. Colloidal forces due to localized singularities and small defect loops

Nematic liquid crystals [1] are characterized by collective and spontaneous orientational order of the long axes of rod-like molecules, which form the nematic liquid crystal. At the same time, there is no positional long-range order of the

centers of gravity of molecules, so a nematic liquid crystal is an orientationally ordered fluid. When in contact with other matter, such as another solid, a liquid, or a gas, the LC molecules at the interface usually interact with the surrounding material. This interaction dictates the orientational (and also positional) ordering of LC molecules at the interface.

The LC–surface interaction has a profound effect, when a foreign particle (such as a microsphere) is immersed into the nematic liquid crystal [7–11]. As the particle forces the LC molecules to align locally with respect to its curved surface, it is obvious that it is not possible to fill the space around the particle with uniformly aligned molecules. In fact, such a curved surface creates frustration for the orientational order of LC molecules, as it has to be curved around a closed surface of the inclusion and it must be uniform far away from the inclusion, which is the natural state for the nematic LC. This frustration generates topological defects [12], which are regions where the degree of order of the nematic LC is strongly reduced and can therefore be considered as regions of molten crystal. For single inclusions, these defects appear in the form of small points and closed defect loops, which accompany the particle and are an inseparable part of it due to fundamental topological laws. The nature of these defects depends on the type of surface ordering of LC molecules at the particle’s surface and are also called ‘singularities’ of the order parameter [1].

When the surface of the particle imposes perpendicular alignment of LC molecules, two different kinds of defects can be observed, which are shown in figure 1. Figure 1(a) shows a $4.7\ \mu\text{m}$ colloidal particle, which is accompanied

by a point-like defect at the top end of the particle, and is therefore called a topological dipole because of its symmetry properties. Figure 1(b) shows the same particle between crossed polarizers of a polarizing microscope. One can see that there is a huge distortion of the nematic liquid crystal around the particle, which spreads out to macroscopic distances. The reason for this long-range distortion is that any disturbance of the long-range order of the nematic LC is also of long range. Furthermore, the distortion of the orientational field is an elastic distortion and stores the elastic energy. The reason for the elasticity of nematic LCs is the broken continuous rotational symmetry of the isotropic phase at the phase transition into the nematic state.

Using optical polarization microscopy, it is possible to determine lines of alignment of the nematic around the particle and its defect, which are shown in figure 1(c). The topological dipole is therefore formed of a colloidal particle and the point defect, and is similar to the electric dipole. A close look at the point defect shows that it has the hyperbolic structure of the director field and is called a hyperbolic hedgehog [5]. On the other hand, the colloidal particle can be considered as a source of orientational field and is called a radial hedgehog. As they are topologically different, we assign topological charges to each type of defect, similar to electric charges, which are the defects, i.e. sources of the electric field.

Figure 1(d) shows another possible configuration of the colloidal particle and its accompanying defect, which now appears in the form of a small defect ring, encircling the particle at the equator [13–15]. This ‘Saturn ring’ defect can be obtained by opening the point defect into a small ring and carries the same topological charge as the hyperbolic point defect. However, the charge is now distributed along the ring. Similarly to the topological dipole, the Saturn ring configuration is called the topological (or elastic) quadrupole for obvious reasons of symmetry of the director field, shown in figure 1(f).

It is observed in experiments [16–20] that when a pair of colloidal particles is brought into close vicinity, the force acts between the particles. This force is obviously mediated by the deformed structure of the nematic liquid crystal and is therefore called the structural force. It may be repulsive or attractive and is always highly anisotropic. For micrometer-sized particles, this force acts over macroscopic separations and is extremely strong. The reason for the onset of this force is understood by considering the elastic deformation of the director field around each particle. When the elastically distorted regions of neighboring particles start to overlap significantly, the free energy of the pair depends on the separation between the particles. This means that a force acts between the two particles, which is due to the topological properties of the director field.

Figure 2 shows the measured separation dependence of the structural force between two collinear and parallel topological dipoles (figure 2(a)) and two topological quadrupoles (figure 2(b)). Here we have used $2.3\ \mu\text{m}$ silica particles with perpendicular anchoring dispersed in the 5CB nematic LC placed in the planar cell. The thickness of the cell was $\sim 8\ \mu\text{m}$ in the case of dipoles and $\sim 4.5\ \mu\text{m}$ in the case of quadrupoles.

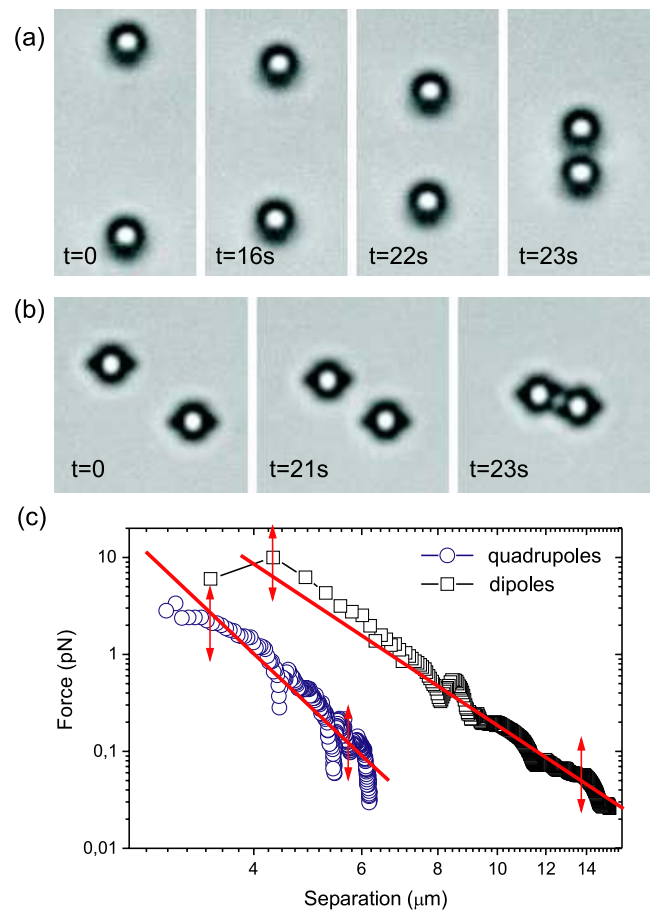


Figure 2. (a) Selected images from the time sequence showing the interaction of two collinear topological dipoles, pointing in the same direction. (b) The interaction of two topological quadrupoles. (c) The pair-interaction force for topological dipoles and quadrupoles. The solid lines are the best fit to the power-law dependence in the marked interval.

The attractive structural force between two colloids is equal to the viscous force on both particles and is determined by measuring the velocity of both particles during attraction [16] and by measuring the viscosity coefficient by observing the thermal motion of a single colloid [21]. Such experiments are very accurate since there are no external electrical or optical fields which can disturb the director field. The force between two elastic dipoles shows a power-law dependence with an exponent of -4 (figure 2(c)), which is similar to the power-law dependence of the electric force between two electric dipoles. A similar situation is obtained for the interaction of topological quadrupoles, where the structural force shows a power-law decay with an exponent of -6 , which is reminiscent of the electric quadrupoles case. This was the basis of the formulation of mean-field theory, which is analogous to the electrostatic interaction between electric multipoles [4]. However, it was recently shown by Pergamenschik and Uzunova [22] that this approach is valid only for spherical colloids with homogeneous anchoring, while in the general case of elastic multipoles more parameters are necessary for describing general multipole deformation of the nematic director field.

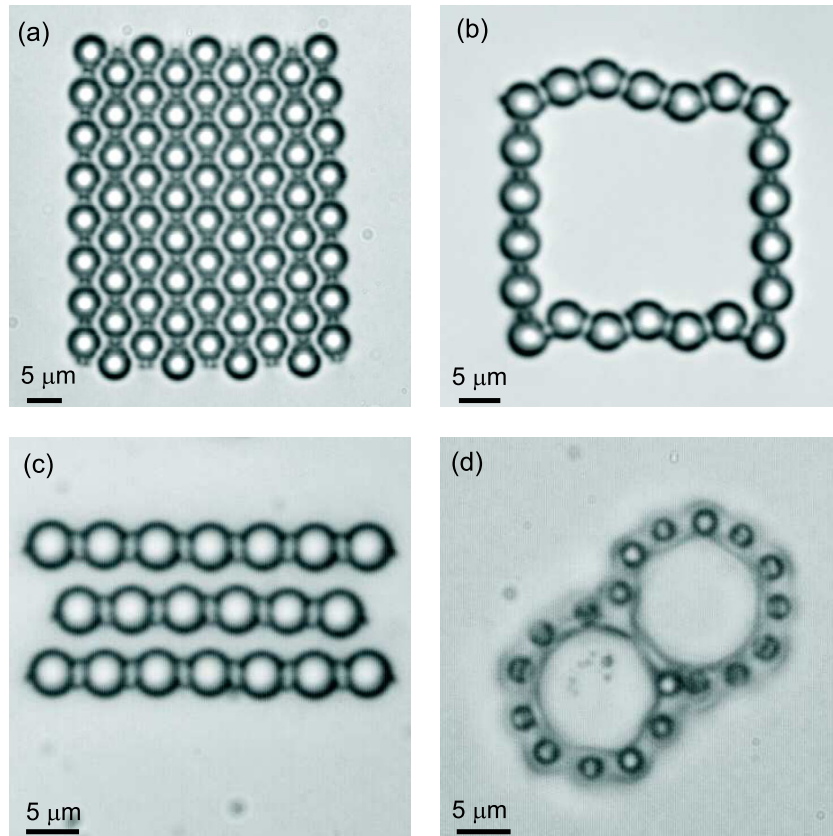


Figure 3. Nematic colloidal structures and superstructures. (a) A 2D nematic colloidal crystal formed from topological dipoles. (b) A colloidal superstructure, assembled from mixed, i.e. dipolar and quadrupolar, colloidal particles. (c) Entangled colloidal wires do not form 2D crystals. (d) The closed defect loop of the ‘figure of 8’ entangled colloidal pair is filled with smaller colloidal particles.

2.2. 2D colloidal crystals and superstructures in nematic colloids

The structural forces in liquid crystals are extremely strong, anisotropic and long range, and have been used for self-assembly and directed assembly of a broad variety of colloidal superstructures in liquid crystals. Nematic 2D colloidal crystals were presented on LC–air interfaces using glycerol droplets as colloids [23, 24]. Several other ordered structures have been presented, like those obtained during cooling from an isotropic phase using a nematic–isotropic interface [25, 26], anisotropic colloidal clusters obtained using laser tweezers manipulation [17] and parallel chain structures obtained after quenching from an isotropic phase [27]. It has also been shown that in a thin nematic layer, 2D colloidal crystals could be assembled from dipolar and quadrupolar nematic colloids [28] and a broad variety of 2D nematic colloidal crystals could be formed in a binary mixture of dipolar and quadrupolar nematic colloids [29, 30]. Furthermore, interesting colloidal superstructures could be assembled in the mixtures of large and small colloidal particles [31]. In all cases, the colloidal binding energy is several orders of magnitude stronger compared to that for water based colloids, and could provide the assembly of nanometer-sized colloidal particles of various shapes. Selected examples of colloidal structures and superstructures are shown in figure 3.

Whereas the experiments on directed assembly of nematic colloids have been performed on micrometer-sized particles,

the question remains as to what the lower limit of colloidal size that could still result in thermally stable colloidal structures and superstructures is. Numerical calculations predict that pair colloidal interaction of the order of $\sim k_B T$ could still be achieved for colloidal sizes of the order of 10 nm [32]. Recent experiments on ~ 100 nm colloidal particles do indeed indicate pair interactions of the order of several $100k_B T$, which is in agreement with theoretical predictions [33]. This means that colloidal structures and superstructures with particle sizes and periodicity of the order of ~ 100 nm and less could be thermally stable in the nematic LC, which makes this class of materials interesting for photonic applications.

2.3. Entanglement of nematic colloids

It was predicted from the numerical simulation studies of Guzman *et al* [34], Araki and Tanaka [32], and Žumer *et al* [35] that another type of colloidal binding should be observed in the experiments, where the topological defect lines entangle two or many colloidal particles in the nematic LC. However, in the experiments with colloidal particles in homogeneously aligned measuring cells, no such regular structure was ever observed. Nearly simultaneously with the work of Araki and Tanaka, the numerical studies of Ravnik and Žumer indicated that such an entangled colloidal state could be realized by quenching the nematic colloids from the isotropic phase throughout the isotropic–nematic phase transition. Whereas

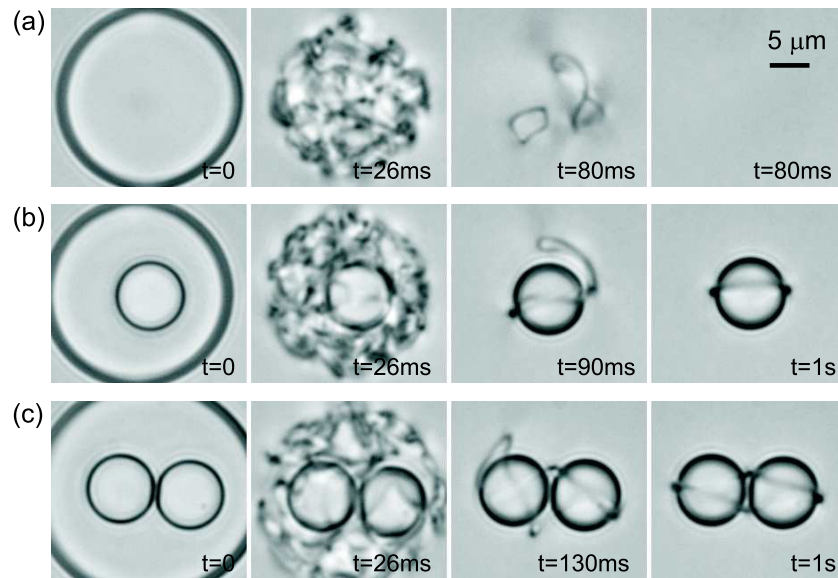


Figure 4. Quenching experiments on nematic colloids. (a) Using high-power laser tweezers, nematic LC is locally heated into the isotropic phase. After switching off the light, the isotropic phase is rapidly cooled into the nematic phase. As a result, a dense network of defect loops is created, which all annihilate. (b) If there is a colloidal particle inside the isotropic melt, all defect loops annihilate except one, which remains pinned to the particle. (c) With two colloidal particles inside the isotropic melt, there is a large probability that after the quench, these two particles are entangled by the ‘figure of 8’ defect loop.

the first quenching experiments were only numerical, using the Landau–de Gennes mean-field theory, it was soon shown in real experiments [36] that entangled colloidal structures could indeed be realized. The experiments were performed by heating locally the nematic LC via heating thin indium tin oxide layer on glass substrates, using a high-power beam of laser tweezers. In this way, the nematic LC was made locally molten, so a micrometer-sized droplet of the isotropic phase was created inside the nematic phase. When the light was switched off, the nematic LC rapidly quenched throughout the isotropic–nematic phase transition. During this quench, a large number of defect lines were created in the form of closed loops, which is shown in the sequence of images in figure 4(a). Finally, after a short period of time, all disclination loops annihilated during the coarsening process and a uniform texture of the nematic LC was obtained. The situation is quite different when the experiment is performed with one colloidal particle present in the nematic LC. As can be seen from the sequence of images in figure 4(b), the coarsening process ends with a single loop encircling the colloidal particle, whereas all other loops annihilate. The reason for this lies in the conservation law for the topological charge, which requires that a defect carrying a charge which is opposite to the charge of the radial hedgehog, carried by the colloidal particle, is created. Finally, topological entanglement of a pair of colloidal particles in the nematic LC is obtained, when a colloidal pair is quenched throughout the isotropic–nematic phase transition. This is shown in the sequence of images in figure 4(c), where the final state is the ‘figure of 8’ entangled colloidal pair. Here, a single defect loop entangles both particles and twists around them. Two other entangled states were also observed, which are called ‘figure of omega’ and ‘entangled point defect’ structure. Some of these structures are chiral and all of them

can extend over an arbitrary number of colloidal particles, forming colloidal ‘wires’.

It is interesting that no periodic and regular 2D entangled colloidal structures were observed in the experiments on homogeneously aligned measuring cells with nematic colloids, although numerical simulations indicated that such structures should exist. It was recently found that 2D entangled colloidal structures do exist in chiral nematic LCs. Here, a rich variety of entangled structures were observed, where the topological defect loops form links and knots [37].

3. Inverted nematic dispersions: a new route to tunable optical microcavities

Dispersions of small droplets of a nematic liquid crystal in a polymer carrier (also called inverted nematic dispersions, or polymer dispersed liquid crystals) have been the subject of very intensive research in the past. The interest in polymer dispersed liquid crystals (PDLCs) was driven by the potential for applications of this material in large-scale privacy windows and displays. When small droplets of a nematic liquid crystal with the diameter comparable to the wavelength of visible light are dispersed in a polymer, one obtains a milky-like dispersion, which strongly scatters light. The scattering is due to the mismatch of the refractive index of the polymer with the refractive indices and the structure of the nematic LC inside the droplet. This scattering of light is strongly reduced when an external electric field is applied to the material. The dispersion becomes transparent, because LC molecules inside the droplets are aligned collectively along the field direction. This is accompanied by a reduction of index mismatch, if the material properties are properly designed.

Instead of considering billions of nematic droplets, which are important for scattering of light in PDLCs, we have

recently considered the optical properties of a single nematic droplet [38]. The internal structure of the director field inside the droplet depends on the anchoring and alignment of LC molecules at the interface to the external polymer material. We have selected a carrier polymer, which induces perpendicular alignment of LC molecules at the interface and has a refractive index which is smaller than the two refractive indices (ordinary and extraordinary) of the nematic LC.

Now, such a micrometer diameter LC droplet can be considered as an optical microcavity [39]. The microcavities are micrometer-sized dielectric objects, which are able to confine light. Confinement of light can be achieved by two different mechanisms: (i) confinement by total internal reflection (TIR) at the interface of the cavity, and (ii) confinement by Bragg reflection at the interface between the cavity and the external Bragg reflector. Both mechanisms provide strong and efficient confinement, where the TIR confinement is broadband and effective over a large frequency range, whereas Bragg confinement is inherently narrow band and frequency selective.

Figure 5(a) shows the principle of light confinement by TIR in an optical microcavity. In the ray picture, light can be considered circulating inside the cavity by subsequent total reflections at the interface. The condition of resonance is met when the light reaches the point of origin with the same phase after one circulation. These resonant waves are also called whispering gallery modes (WGMs) after the famous acoustic effect in spherical domes. In the wave picture, one considers the electric field distribution inside the cavity, and the resonant modes are characterized by three mode numbers. Figure 5(b) shows a micrograph of a $\sim 15 \mu\text{m}$ diameter droplet of the nematic liquid crystal 5CB (pentylcyanobiphenyl) in a PDMS polymer. The structure of the nematic liquid crystal inside the droplet is radial, and the molecules are directed along the radius, thus pointing from the center of the droplet towards the LC/PDMS interface.

Such a radial nematic microcavity does indeed support optical WGM modes, as has been shown recently [38]. The modes are quite narrow (tens of GHz), indicating rather high quality factors ($\sim 10,000$) of the LC microresonators. Most important, these modes can be tuned by an external electric field. Namely, the application of the external electric field forces the molecules to align collectively into the field direction. This changes the refractive index for light circulating along the droplet curved interface, which also changes the condition for resonance, which is then fulfilled for another wavelength. As a result, the WGMs shift with the applied electric field, as presented in figure 5(c). This shift is huge even for moderate values of the electric field, which makes radial nematic LC microresonators potentially interesting for applications in photonic devices. Tunable microcavities are at the heart of important photonic devices, such as optical switches, optical spatial multiplexors (re-directors) of optical signals and tunable lasers. They could be basic tunable optical resonant elements for future technological platforms for soft matter photonic devices. We should note that another class of LC microresonators, i.e. Bragg resonators, based on cholesteric liquid crystals confined to small droplets, could

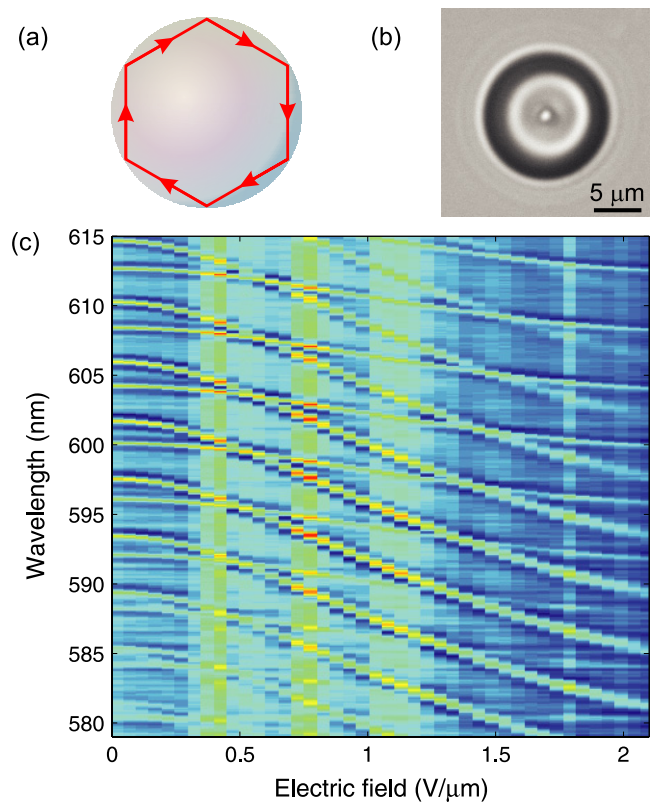


Figure 5. (a) In an optical spherical microcavity, light can be considered circulating inside the microsphere by subsequent total internal reflection. (b) A microscope image of a microdroplet of a nematic liquid crystal in a polymer. (c) The spectrum of light emitted from the microdroplet of a nematic LC exhibits sharp lines, which are the WGMs. After the application of an external ac electric field, the positions of the WGMs shift strongly.

form a very powerful basis for tunable microlasers. This idea was first proposed by Mušević and can be found in the supplementary material of [38]. The operation of the first dye-doped microlaser based on a Bragg resonator formed from a cholesteric LC droplet was just recently demonstrated successfully [40].

4. Conclusions

The aim of this paper was to review the state of the art and very recent developments in the rapidly developing field of nematic colloids. In just a few years this field has evolved from a set of scattered experiments and ideas into a coherent field, which has revealed not only a rich variety of fundamental phenomena, but has also shown a great potential for applications in soft matter photonic materials. Namely, we have shown that there are a number of different mechanisms in nematic LCs which are able to bind microparticles with energies several order of magnitude larger than $k_B T$. Furthermore, we have shown that one could produce LC based tunable microresonators and also tunable lasers. Now, it is straightforward to conclude that in principle, one should be able to combine binding mechanisms and interparticle structural forces with active properties of LC microresonators and microlasers to assemble stable soft matter photonic microcircuits. Although this is highly speculative,

we anticipate that many interesting micro-optical phenomena could emerge within photonics, based on soft matter.

Acknowledgments

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