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Forces in nematic liquid crystals: from nanoscale interfacial forces to long-range forces in nematic colloids

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INVITED ARTICLE

Forces in nematic liquid crystals: from nanoscale interfacial forces to long-range forces in nematic colloids

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In this paper we give an overview of experiments that provided an insight into the nature of forces between surfaces and objects in a nematic liquid crystal. These forces, also called ‘structural forces’, are the consequence of the long-range orientational order and orientational elasticity of nematic liquid crystals. Owing to their fundamental as well as technological importance, forces between objects in liquid crystals have been a subject of growing interest during the last decade. Experimental observations and studies of structural forces are described from nanoscale interfacial forces, measured by an atomic force microscope, to the micro-scale forces between colloidal particles in nematics, studied by laser tweezers and optical video microscopy.

Keywords: forces in liquid crystals; interfacial forces; nematic colloids

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1. Introduction

Owing to their long-range orientational order, liquid crystals transmit static torques for the same reason that solid crystals are able to transmit static forces (*1*). This field has been left unexplored for a long time, because of the lack of appropriate experimental techniques that would enable one to measure very small forces between microscopic objects with nanometre to micrometre separation between them. With the advent of novel experimental techniques, such as atomic force microscopy (AFM) (*2*) and laser tweezers (*3*), it has become possible to do this.

This work was initiated a long time ago by very simple arguments, which can be expressed briefly as follows. Whenever there is a wall or a surface of arbitrary shape inside the liquid crystal (see Figure 1), it breaks the continuous translational symmetry of a liquid crystal and induces changes in the orientational and positional order of a liquid crystal in the vicinity of the wall. If the second wall is brought in close proximity to the first wall, the influence of both walls will start to overlap. This will make the total free energy of the liquid crystal dependent on the separation between the walls. In terms of physics, this separation dependence of the free energy of the confined liquid crystal will give rise to the force F between the two walls. These forces are named ‘interfacial structural forces’, which is an expression well known from the physics of water colloidal dispersions (*4*).

When considering the nature of these forces, they are obviously related to the nature of the ordering of liquid crystalline molecules in the interfacial region close to the wall. This means that they imply both orientational (i.e. nematic) order and smectic order (i.e. layering), which are both a direct consequence of molecular anchoring at the interface. In principle one could therefore obtain very valuable information about the degree of the orientational and smectic order in a very thin layer of a liquid crystal, confined between two flat (for convenience of calculations) surfaces. Furthermore, one could test the validity Landau–de Gennes (LdG) theory at the nanoscale, by comparing theoretically predicted structural forces with those that were measured. One could therefore answer the question whether LdG theory is valid for liquid crystal layers that are only several molecules thick.

If we now consider interfacial forces on a much larger scale, say micrometre wall-to-wall separation, we note that an inclusion in a liquid crystal (such as spherical colloidal particles in Figure 2), creates an elastic disturbance in the local orientation of the nematic liquid crystal around it, because of the interaction of its curved surface with the nematic molecules. Now, if we bring another microscopic inclusion into close vicinity with the first, this will

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Professor Musevic received the 2008 Samsung Mid-Career award for his work on the physical studies of antiferroelectric and ferroelectric liquid crystals, measurements of interfacial forces, colloidal particles and their manipulation in nematic liquid crystals.

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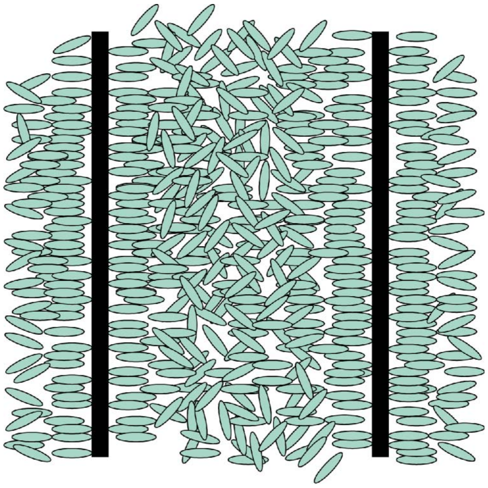

Figure 1. Walls in a liquid crystal induce changes of the liquid crystal orientation in their vicinity, which makes the free energy of the system dependent on the separation between the walls. This is the reason why forces between surfaces appear in liquid crystals.

inevitably create forces between the two particles, because of the overlapping of the distorted liquid crystal layer around each inclusion, which again makes the free energy of the system dependent on the particle separation.

It is important to note that inclusions in liquid crystals are specific and behave quite differently to colloidal particles in isotropic solvents, such as water. When studying the behaviour of inclusions of particles in nematic liquid crystals, we meet the concept of topological charge and its conservation laws (*5*), which have deep implications for the nature of nematic colloidal interactions. As a result of conservation of the topological charge, defects always accompany inclusions, and the disturbance of the director field around the inclusion is spatially strongly anisotropic even though the colloidal particles are spherical. When several colloidal particles are brought together in the nematic liquid crystal, the total energy of the system depends on particle separation and their relative orientation, which means that a force of structural origin emerges between the particles. As these structural forces are due to the collective order in liquid crystals, they are anisotropic and of long range, therefore reflecting the nature of the orientational order in liquid crystals.

This paper is organised into two separate parts. In the first part, we describe our AFM experiments on interfacial forces in nematic liquid crystals, therefore dealing with the fine details of the nature of ordering of liquid crystal molecules at very small nanometre separations from the confining wall. The second part is devoted to the 'far field' picture, i.e. the interactions and forces between objects in nematic liquid crystals, which present some most fascinating examples of self-assembly in complex matter.

2. Interfacial forces in liquid crystals

The structure of liquid crystals at interfaces has been studied in the past using conventional techniques such as nuclear magnetic resonance (NMR), quasielastic light scattering and X-rays. As the proportion of liquid crystal in the interfacial region relative to the bulk is very small, the signal contribution from the bulk dominates. To overcome this problem and increase the surface-to-volume ratio, liquid crystals have been introduced into porous media, such as aerogels or anapore membranes, have been filled with granular materials, such as silica spheres and

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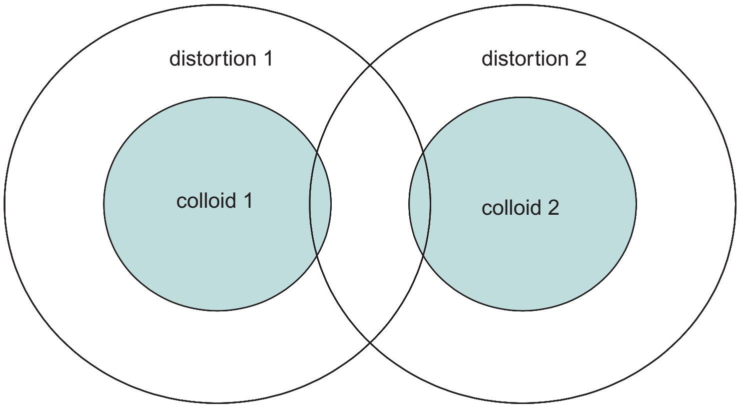

Figure 2. When colloidal particles are inserted into a liquid crystal, they create regions of elastic distortion around them, spreading out to micrometre separations. The free energy of a liquid crystal with inclusions depends on the separation between the particles, which is the reason for the appearance of forces between particles in nematic colloids.

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nanoparticles, or have been dispersed in polymers and water. Later, new surface-sensitive experimental methods have been introduced into the study of liquid crystal interface, such as the second harmonic and frequency sum generation, ellipsometry, surface force apparatus (SFA), AFM and soft X-ray absorption spectroscopy (NEXAFS). A general overview of the literature can be found in (*6*).

The structure of a liquid crystal at a solid interface can be well described using LdG theory, which describes the molecular order on the mesoscopic scale. In fact, LdG theory is naturally suited for the description of structural forces, as it is based on the expansion of the free energy of the system in terms of an (or several) order parameter, which describes the ordering. As the structural force between bodies $\mathcal{F}$ is a negative derivative of the free energy F with respect to the separation d, $\mathcal{F} = -\partial F/\partial d$, LdG theory fits perfectly into the problem of the mathematical description of structural forces in liquid crystals. Indeed, LdG theory has been used to predict different types of liquid crystal interactions before they have actually been observed. For example, mean field nematic structural forces (i.e. when the fluctuation effects are neglected) were considered by a number of authors (*7–11*), and many interesting predictions were made, including surface-induced capillary condensation of the nematic phase (*7–8*) and pseudo-Casimir effect in liquid crystals, where the interaction is generated by fluctuations of the liquid crystalline order (*12*). The pioneering experimental work on structural forces in the nematic liquid crystals has been done by Horn *et al.* (*13*) using a SFA, which was later continued by a number of authors (*14–17*). A lot of work on lyotropic liquid crystals has been done on presmectic interaction (*16*, *17*) and capillary condensation of lamellar lyotropic phases (*18–19*).

Here, we describe our experiments, where we have used temperature-controlled AFM (*20*, *21*) to study the forces, mediated by a very thin layer of the nematic or smectic liquid crystal, confined between a micrometre-sized sphere (attached to the AFM cantilever) and a flat surface. Owing to the experimental limitations when using optical detection of forces with AFM, most of the experiments on structural forces have been limited to the temperature region around the isotropic–nematic phase transition, where the light scattering by liquid crystal is small. On the other hand, this region is very interesting, because the free energy of the liquid crystal close to the phase transition depends strongly on external influences, such as the surface anchoring. In the vicinity of the phase transition, the systems are usually soft and very susceptible to external action, which means that one should expect very large interfacial forces in this region. This enhancement of forces close to the phase transition is therefore an advantage and could give us detailed insight into the interfacial properties of a liquid crystal.

Let us consider qualitatively what kind of forces are expected, when a nematic liquid crystal is confined between two closely positioned flat surfaces with equal (let us say homeotropic) surface anchoring, and the temperature is slightly above the clearing point T_{IN}. We know that an orienting flat surface will induce in the isotropic phase a small degree of the orientational order S, which will decay exponentially from each surface. Now, if we consider the situation, shown in Figure 3, we see that the ordered regions emanating from each surface start to overlap when the wall separation is smaller than the correlation length. This situation of 'sharing' regions of order is, qualitatively speaking, in favour of both surfaces and lowers the total free energy of the system. This reduction of the free energy will be even larger if the surfaces are closer. This leads us to a conclusion that there should be an attractive pre-nematic structural force between two equal surfaces immersed in a nematic liquid crystal at a temperature slightly above the T_{IN}.

The surface-induced pre-nematic force was actually observed in our experiments with 5CB nematic liquid crystal on silanated surfaces inducing strong homeotropic alignment (*22*) and is shown in Figure 4. The force is attractive, as expected for a pre-nematic force, and it is very small. The magnitude of the measured force, normalised to the radius of the microsphere, $\mathcal{F}R$, is typically $6 \times 10^{-6} Nm^{-1}$, which for $2R = 7\ \mu$m corresponds to a force of about 20 pN. This attractive force can be well described as a sum of an attractive pre-nematic force and usual van der Waals attraction across the liquid crystal medium (*22*).

However, it is known that a solid surface induces not only orientation of neighbouring liquid crystal molecules, but also induces positional order in the

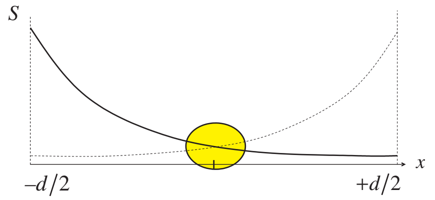

Figure 3. Origin of pre-nematic structural force. Schematics of the separation dependence of the nematic order parameter S between two orienting walls separated by d. The temperature is slightly above the T_{IN}, and each wall induces a small pre-transitional nematic order, which decays within a correlation length from each surface. Note the region of overlapping of ordered regions, emanating from each surface.

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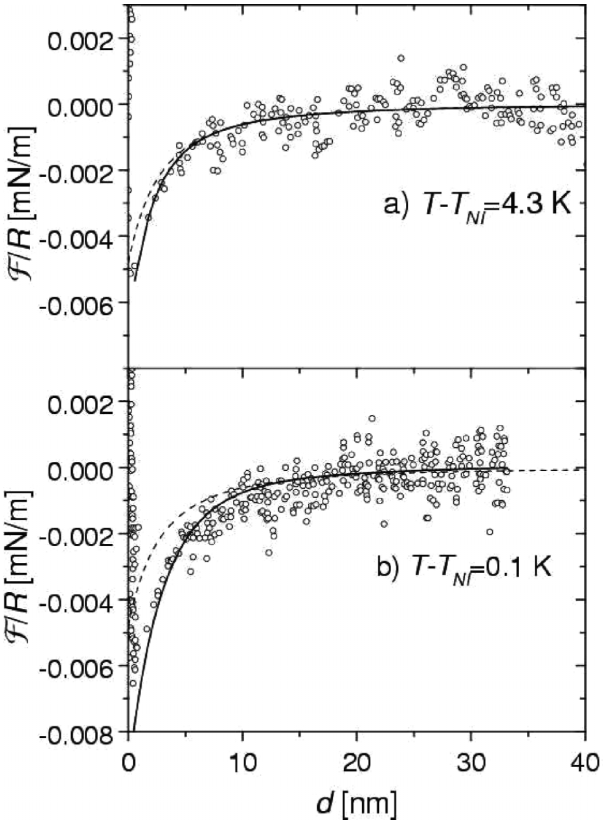

Figure 4. Attractive force $\mathcal{F}$ between a $2R = 7\ \mu$m silanated glass sphere and silanated flat glass surface in the presence of 5CB. The force is normalised to R. The temperatures are just slightly above the phase transition temperature T_{NI}. Reproduced with permission from (22).

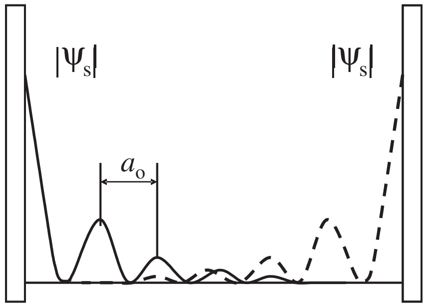

Figure 5. Overlapping of the two pre-smectic ordering profiles from each of the confining surfaces leads to an oscillatory pre-smectic force between the two surfaces.

first several layers adjacent to the solid wall. For silanated surfaces, this positional anchoring can be so strong, that the first layer of n-CB nematics is actually a smectic-like monolayer, as was shown in our first AFM experiments (*23*). On top of this 'first', smectic-like layer, there is the liquid crystal interface with a small, surface-induced smectic order, which decays as we move into the nematic or isotropic bulk. This surface-induced smectic ordering gives rise to the pre-smectic forces in the same way as it induces the pre-nematic forces. The pre-smectic force between two equal surfaces was first considered by de Gennes (*10*), and can be qualitatively understood using simple arguments, as illustrated in Figure 5. This figure shows the dependence of the smectic order parameter $\Psi(x)$ on the separation from the confining wall. It has the highest value Ψ_s at the confining surface and then decays into the bulk, because it is not as stable as the bulk solution. The spatial period of this oscillating pre-smectic ordering profile is equal to the separation between the two consecutive smectic layers. Now, when the separation between the two confining surfaces is such that the maxima of the pre-smectic ordering originating from the left surface in Figure 5 matches the maxima of the pre-smectic profile from the right surface, the total free energy will be low, because the surfaces can share equally preferred regions of smectic order. However, if the maxima matches the minima, the total free energy will be high, because the situation is frustrated for both surfaces. This means that the total free energy now depends periodically on the separation between the two confining surfaces and the resulting pre-smectic force is expected to be oscillatory as well.

The oscillatory pre-smectic force was indeed observed in AFM experiments by Kočevar and Muševič (*24*) and is shown in Figure 6 for different temperatures just above the isotropic–nematic transition point in 8CB. The force is relatively strong and easily observed and could be perfectly described by de Gennes' calculations (*24*), as is evident from the solid lines, representing the theoretical prediction of de Gennes. Moreover, it is evident that de Gennes' description is in perfect agreement with the experiment down to the length-scale of several nanometres. This proves the inherent strength of the mean-field theory of de Gennes.

There are other examples of structural forces in nematic liquid crystals, such as the capillary condensation of the nematic phase out of the isotropic phase, confined in a narrow gap between two strongly orienting surfaces. The effect was first predicted by P. Sheng in 1976 (*7*). It is conceptually similar to the condensation of water in pores and cracks with hydrophilic surfaces. Owing to the surface field, condensation of water is induced in a tight gap in the form of a capillary water meniscus. This meniscus is curved due to the surface tension of the water–vapour interface, and as a result of the change of pressure across this meniscus, a strong attractive force acts between the two surfaces. The phenomenon has been known for quite a long time and was explained by Lord Kelvin in 1871. In the isotropic phase of the nematic liquid crystal, capillary condensation of the nematic phase occurs when the

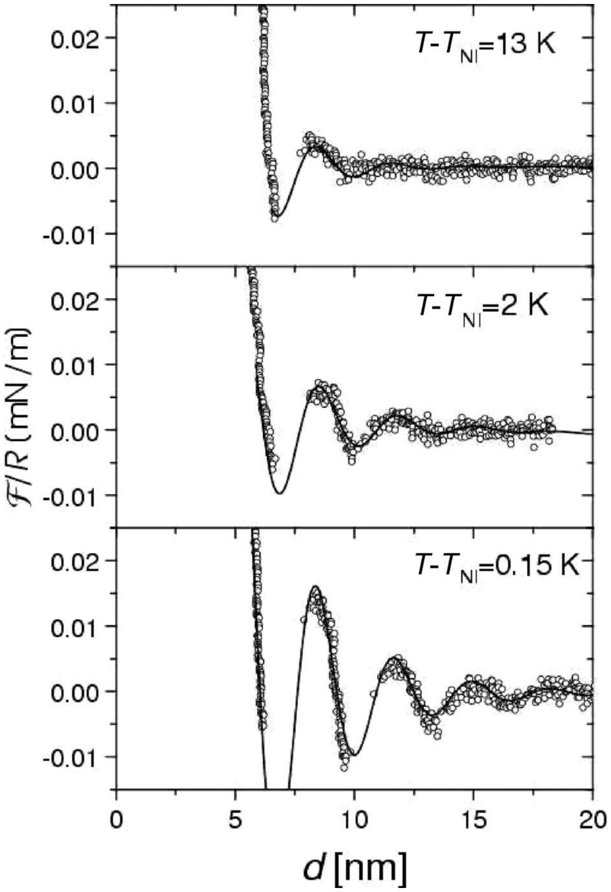

Figure 6. Pre-smectic force in the isotropic phase of 8CB nematic liquid crystal between two DMOAP silanated glass surfaces. Courtesy of K. Kočevar. Reproduced with permission from (24). Copyright 2006, The American Association for the Advancement of Science.

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surfaces induce a large degree of orientational order. In this case, a very large spatial gradient of the nematic order is created in the isotropic phase, which is energetically unfavourable. At some separation, it is more favourable to phase-transform the isotropic phase of the liquid crystal into the ordered, nematic state, which occurs in the form of a capillary bridge of a nematic phase, condensed in the small gap between the two confining surfaces. This bridge creates an enormous force due to the nematic–isotropic interface, separating the isotropic and nematic phase. The observation of the capillary condensation was reported in an AFM experiment by Kočevar *et al.* in 2001 (*25*, *26*).

3. Interactions in nematic colloids

Nematic colloids were first used for 'decorating' liquid crystalline surfaces, where it was observed that gas bubbles at the nematic–air interface formed chains, aligned along the nematic director (*1*, *27*). Whereas in those early experiments, the nematic–air surface tension played an important role in the organisation of bubbles into chains at the nematic–air interface, this effect has been removed in the experiments on nematic emulsions by Poulin *et al.* (*28*, *29*). They have shown that water droplets, dispersed in the nematic liquid crystal, did not coalesce, but formed long chains, where each droplet in a chain was separated by a topological defect, preventing their contact. It soon became clear that the formation of colloidal chains is due to the elastic deformation of the director field around each colloidal particle and sharing of regions of equal distortions is the mechanism, responsible for these *structural forces* in nematic colloids. It was later shown that these structural forces in nematic colloids are long range, with a typical binding energy of several $1000k_BT$ per particle (*31–35*) and give rise to some fascinating one- and two-dimensional self-assembled colloidal structures (*36–41*).

The binding energy of several $1000k_BT$ per colloidal particle of micrometre size is typically thousands of times stronger compared with the binding in water-based colloids of comparable size, where the colloidal interactions are spatially isotropic and are controlled by the fine balance between the attractive van der Waals dispersion force and the repulsive electrostatic and steric forces. The extremely strong and anisotropic binding of nematic colloids by the structural forces makes these systems technologically very promising for future applications of colloids in photonic devices.

The origin of structural forces in nematic colloids can be understood in a similar way as the interfacial forces in nematics, discussed in previous section. When two colloidal objects are brought close together in the nematic phase, the regions of their elastic deformation start to overlap, which means that the free energy of the colloidal pair depends on the separation between the two particles. This is the reason why the force of structural origin appears between the particles, which is either attractive or repelling, but is of long (i.e. micrometre) range, because of the long range of distortion of the nematic director field around the colloidal inclusion. It has been shown by Lubensky *et al.* (*34*), that the concept of topological charge (*42*) plays an important role in understanding the nematic colloidal interactions, which could be described within a mathematical framework, similar to the electrostatic interaction between electrically charged objects.

When a small spherical particle, which has been surface-treated to adopt homeotropic alignment of the nematic liquid crystal such as that shown in Figure 7(a), is immersed in the nematic liquid crystal, the resulting elastic distortion of the liquid crystal around the particle can be either of dipolar or quadrupolar symmetry,

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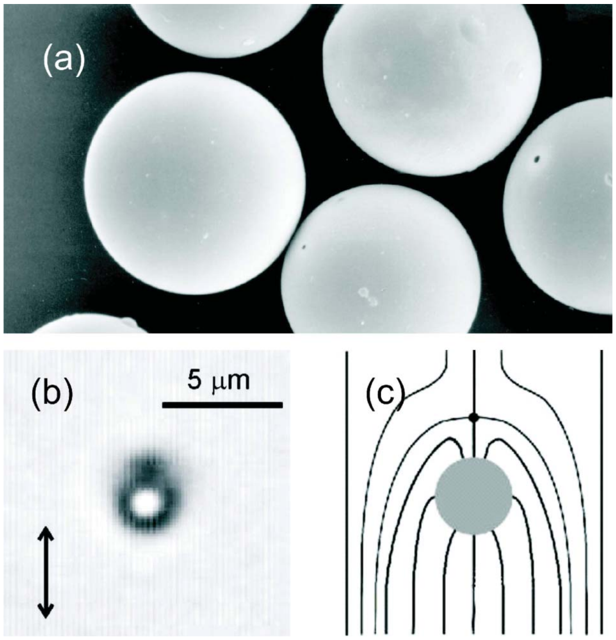

Figure 7. Dipolar nematic colloidal particles. (a) Glass microspheres, treated to induce homeotropic nematic alignment at their curved surfaces. (b) Micrograph of a dipolar nematic colloidal particle with characteristic point defect. (c) Schematics of the director field of a dipolar nematic colloid. Reproduced with permission from (*36*). Copyright 2006, The American Association for the Advancement of Science.

illustrated in Figures 7 and 8. The dipolar colloidal particle is accompanied by a dark spot (Figure 7(b)), which is a *hedgehog point defect*, located along the overall nematic orientation (*42*). Together with the colloidal particle itself, it forms an object of dipolar symmetry. Under certain circumstances (such as small diameter of the particle, or external fields (*43*)), the point hedgehog defect can open into a small defect ring, encircling the colloidal particle. It was first observed by Yuedong and Abbott (*44*) and was called a *Saturn ring defect*. Unlike dipolar nematic colloids, this configuration has a quadrupolar symmetry and interacts quite differently with other colloidal particles.

When two dipolar nematic colloidal particles are placed in close vicinity, let us say several micrometres, they usually interact very strongly. The interaction can be either repulsive or attractive, depending on the relative orientation of their topological dipoles. When placed collinearly along the overall nematic orientation with their dipoles pointing in the same direction, they will strongly attract and form long colloidal chains, as illustrated in Figure 9. The binding energy of a dipolar nematic colloidal pair can be as high as several $1000k_BT$ for micrometre-sized particles. Quadrupolar interaction is not as strong as the dipolar, and results in a spontaneous formation of kinked quadrupolar chains (*38*).

It was shown in 2006 by Muševič *et al.* (*36*) that nematic colloids also self-assemble in two dimensions, forming at least two different colloidal lattices of dipolar and quadrupolar particles. Figure 10 shows an example of a two-dimensional dipolar nematic colloidal crystal, which was assembled using the laser tweezers manipulation in planar nematic cells with the thickness somewhat larger than the diameter of the colloidal particles. The crystal can be seen as assembled from chains of dipolar colloids, oriented along the overall nematic orientation. One can also notice that each neighbouring chain is oriented in the opposite direction, as illustrated by coloured arrows on the right image. One can understand the mechanism of two-dimensional self-assembly in terms of a

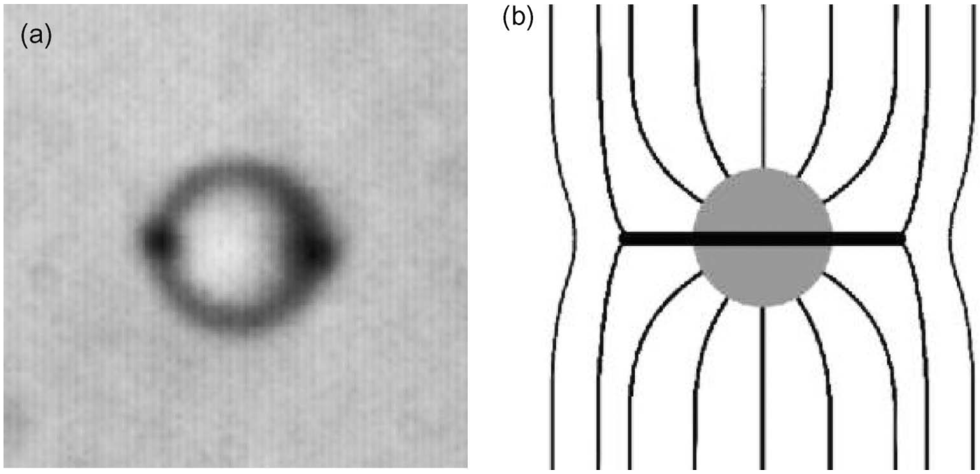

Figure 8. (a) Quadrupolar nematic colloidal particle with the characteristic Saturn ring defect. (b) Schematics of the director field around the quadrupolar nematic colloid.

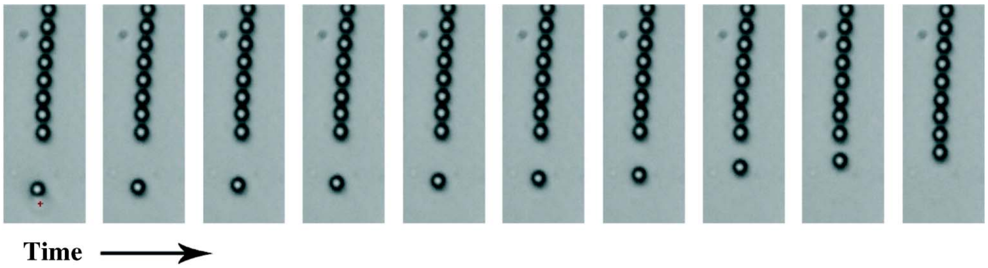

Figure 9. Set of microscope images, showing long-range attraction between a dipolar colloidal particle and a colloidal chain. The dipoles are pointing in the same direction.

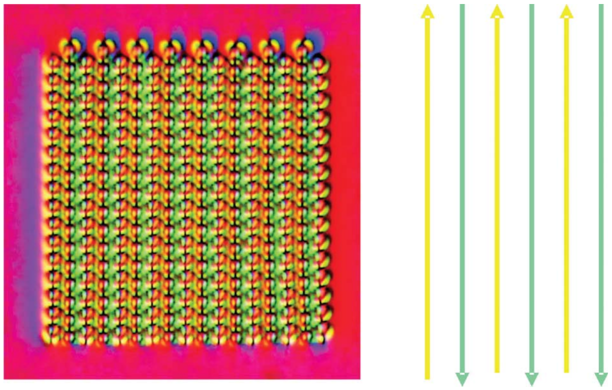

Figure 10. Two-dimensional dipolar nematic colloidal crystal. Reproduced with permission from (*36*). Copyright 2006, The American Association for the Advancement of Science.

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simplified electrostatic picture: each dipolar chain will tend to attract another chain, oriented in the opposite direction. It turns out that this simple electrostatic picture indeed works well in thin planar cells, but the interactions are more complex when one tries to assemble three-dimensional nematic colloidal crystal. This complexity was recently addressed theoretically by Pergamenshchik and Uzunova (*45*), showing that in three dimensions the nematic elastic interaction significantly differs from electrostatics both in mathematical structure and physical implications.

The richness of the family of nematic colloidal crystals has been illustrated recently by Ognysta *et al.* (*40*), who have shown that in a mixture of dipolar and quadrupolar nematic colloids, a whole new family of different colloidal crystals of different symmetry and structures are stable. In this sense, binary mixtures of dipolar and quadrupolar nematic colloids can be considered as a mixture of two different chemical elements 'A' and 'B', which can form different 'chemical bonds' due to the anisotropy of interaction between the dipolar (i.e. element 'A') and quadrupolar (i.e. element 'B') colloids. More than 12 different binary nematic colloidal crystals have been assembled successfully.

Finally, let us mention another fascinating mechanism of nematic colloidal interaction, which was discovered recently and has no analogue in water-based colloids. This is binding via defect rings and loops, entangling two or more colloidal particles, which was predicted several years ago (*46–48*) and first proven in recently published laser tweezers experiments and LdG analysis (*39*). Whereas dipolar and quadrupolar nematic colloids interact by sharing regions of elastic distortion, the mechanism of binding via entangled defect loops and rings is quite different, and can be illustrated in Figure 11.

Here, the liquid crystal was first heated locally into the isotropic phase using absorption of a high-power laser beam, thus creating an island of the isotropic phase, surrounding two colloidal particles and being surrounded by the nematic. After the light was switched off, the isotropic island surrounding both colloidal particles rapidly cooled into the nematic phase, creating a dense tangle of defect lines. In a fraction of a second, the lines annihilated, transforming the defect tangle into well-oriented nematic and finally a single defect line remained, encircling both colloidal particles in the form of a 'figure of eight'. Two other entangled structures were observed experimentally, and the entanglement was not limited just to colloidal pairs; colloidal chains and wires of arbitrary length could also be formed.

It is important to note that the entanglement mechanism is in its nature quite different from binding via sharing of elastically deformed regions, induced by point or small ring singularities. Instead, the binding energy is concentrated here in the formation of defect lines, and therefore a string-like force is expected and also observed between entangled colloidal particles.

4. Conclusions

In this short review we have attempted to describe progress and the state of the art in the field of structural forces in nematic liquid crystals during the last 10 years, emphasising the underlying physical picture and concepts. The review has been concentrated on work

Figure 11. Entanglement of a colloidal pair after quench from the isotropic phase.

performed by our group and therefore many important and valuable references could not be included due to the limited space available. Whereas at the beginning it was not clear whether structural forces are at all observable using the available techniques, it is now evident that one can obtain valuable information about the structure of liquid crystal–solid interfaces with unprecedented accuracy and spatial resolution. In my opinion, one of the most striking result is experimental proof that LdG theory could describe quantitatively the behaviour of confined nematic liquid crystals from micrometre to nanometre separations between surfaces. This gives us confidence that we are able to model and predict theoretically many interesting phenomena and devices (and possibly their performance) at the nanoscale. When going from the nanoscale forces to larger scale and nematic colloids, we see an amazing richness of phenomena related to particle interactions, mediated by the liquid crystal. Most of them have no analogue in ordinary, water-based colloids and this opens a new method to construct novel devices in the future.

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