

# Liquid Crystal Aided Nanotechnology/Nanoscience

Subjects: Physics, Condensed Matter

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The research field of liquid crystals and their applications is recently changing from being largely focused on display applications and optical shutter elements in various fields, to quite novel and diverse applications in the area of nanotechnology and nanoscience. Functional nanoparticles, such as ferroelectric and magnetic particles, have recently been used to a significant extent to modify the physical properties of liquid crystals. Also, intriguing photonic functionalities can be realized by adding nanoparticles such as quantum dots, metal particles, semiconductors, etc. into liquid crystals. The self-organization of liquid crystal molecules is exploited to be used as order templates to orient nanoparticles. Similarly, anisodiametric nanoparticles such as rods, nanotubes and flakes are shown to form lyotropic liquid crystal phases in the presence of isotropic host solvents at a certain concentration.

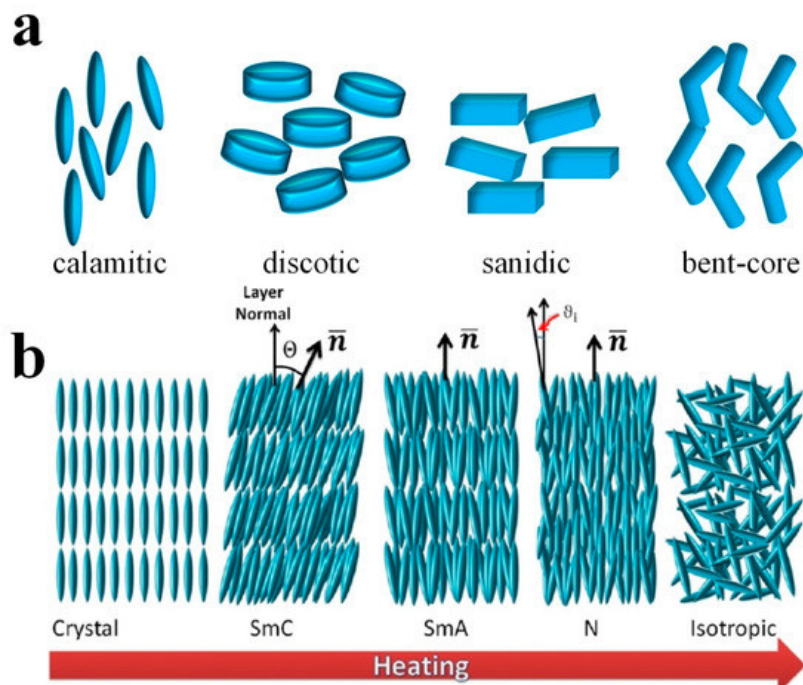
Keywords: Liquid crystals ; ferroelectric nanoparticles ; quantum dots ; graphene oxides

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## 1. Introduction

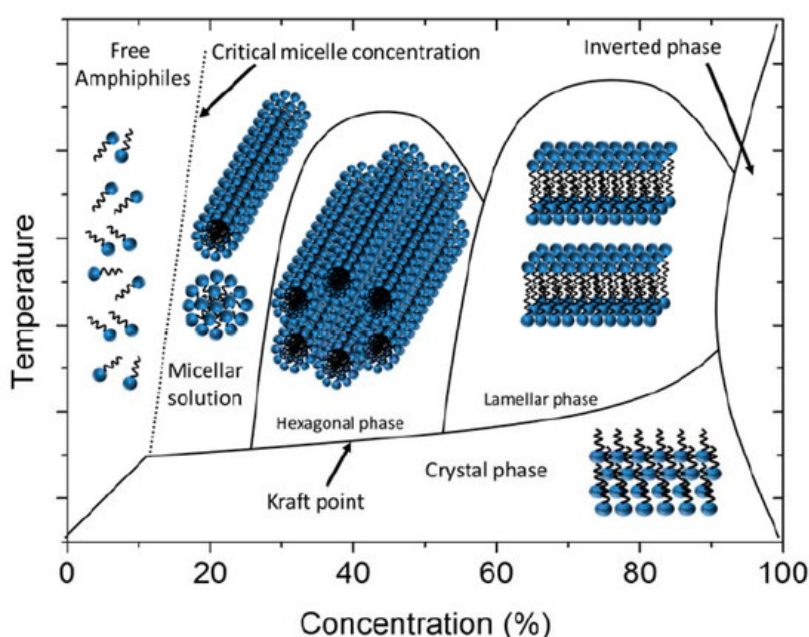
For many materials, the transition between the liquid and the solid phase is not a single-step process, but a range of various mesophases, which are called liquid crystals (LCs). LCs are self-organized anisotropic fluids that are thermodynamically located between the isotropic liquid and the crystalline phase, exhibiting the fluidity of liquids as well as the long-range lattice order that can only be found in crystalline solids <sup>[1][2][3]</sup>. Generally, LCs are composed of anisotropic building blocks (usually of rod or disc shape), which are spontaneously oriented along a specific direction, called the director  $\mathbf{n}$  <sup>[4]</sup>. Without an external alignment force, the director of a nematic LC, the simplest phase of LC, whose molecules are only orientationally ordered, is usually spatially changed continuously but randomly over large spatial extensions (except for defects, where the director may vary suddenly and drastically) <sup>[5][6]</sup>.

Normally, one can split LCs into two typical categories, i.e., thermotropic LCs and lyotropic LCs <sup>[7][8][9][10]</sup>. Thermotropic LCs are usually further distinguished according to the shape of their constituent molecules, being called calamitic for rod-like, discotic for disk-like and sanidic for brick- or lath-like molecules (Figure 1a) <sup>[11]</sup>. A typical calamitic mesogen is generally composed of a rigid core, often incorporating phenyl and biphenyl groups, and two flexible endgroups, often alkyl or alkoxy chains. A common structure of discotic mesogens is a rigid, disk-like core to which six flexible endgroups are attached. Apart from these conventional mesogens, research attention has been recently focused on the so-called non-conventional LCs <sup>[12]</sup>, which are neither rod- nor disk-shaped. Among them, bent-core LCs <sup>[13]</sup>, in particular, have received attention due to their unique effects of the observation of chirality from achiral molecules, resulting from sterically induced packing of the bent-core mesogens <sup>[10]</sup>, such as ferroelectricity or the formation of helical superstructures in the B7 phase <sup>[14]</sup>. Thermotropic LCs are commonly constituted by single organic entities or mixtures thereof, which exhibit various mesophases at different temperatures or pressures <sup>[15]</sup>, illustrated in Figure 1b. As the temperature rises, a typical thermotropic LC passes through higher ordered phases, also called soft crystals, the hexatic smectic phases with positional order as well as bond orientational order, through the fluid smectic phases (SmC and SmA), which exhibit both positional and orientational order, and finally to the nematic phase (N) with purely orientational order, into the isotropic phase. The number of different phases observed depends on the chemical composition, symmetry and order of the LC molecules. About 25 different thermotropic phases are known to date, and they are still increasing in number.

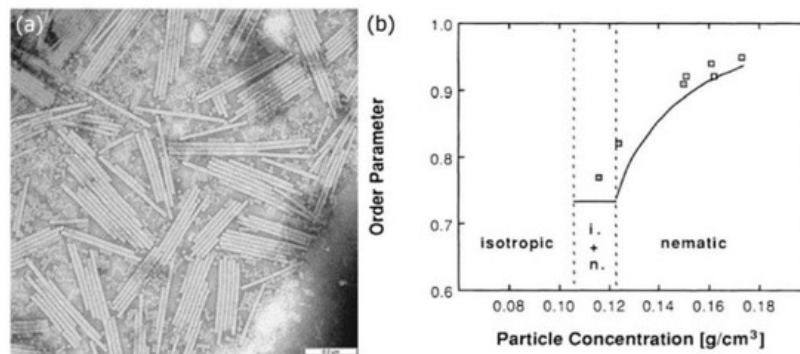


**Figure 1.** (a) The various shapes of LC molecules. (b) A typical illustration of different thermotropic LC phases observed on heating from crystalline state. Reproduced with permission from [2]. Copyright (2017) MDPI.

In contrast, lyotropic LCs consist of at least two different kinds of components: a collection of anisodiametric molecules and particles dispersed in a suitable solvent (Figure 2). These systems will thus always be mixtures, with the main control variable being the concentration. Unlike thermotropics, the phase transitions of lyotropic LCs are not merely dependent on the temperature of the system, but also, mainly, on the relative concentration of each component [15]. Being mixtures, there will always be two-phase regions at the phase boundaries between two different lyotropic phases. One can distinguish lyotropic LCs into three different kinds: (i) amphiphilic lyotropics, (ii) colloidal lyotropics, and (iii) chromonics, where the constituent molecules are dye molecules in a suitable solvent. As the name indicates, amphiphilic lyotropic LCs are usually composed of amphiphilic molecules upon addition of a solvent [16], often water. As Figure 2 shows, with increasing amphiphile concentration, due to the segregation of hydrophobic and hydrophilic regions, these molecules self-assemble into spherical or rod-like micelles, leading to the formation of the hexagonal phase, cubic phase or lamellar phase. Nevertheless, the solute component of a lyotropic LC need not always be molecular in nature, but may also consist of much larger (solid) particles with anisotropic shapes. It can also be of colloidal size [2]. Such materials would then be known as inorganic liquid crystals, LC clays, but also nanotubes, graphene oxide or biological structures such as viruses (Figure 3).



**Figure 2.** A typical lyotropic LC phase diagram of amphiphilic molecules dissolved in a solvent. Cubic phases may be observed at different positions in the phase diagram. At very high concentrations, the inverse phases are located. Reproduced with permission from [2]. Copyright (2017) MDPI.



**Figure 3.** (a) SEM image of a collection of tobacco mosaic virus particles. Scale bar: 0.2  $\mu\text{m}$ . Reproduced from Wikimedia Commons, with no author name supplied. (b) The orientational order parameter of the viruses increases as their concentration in the solvent is increased. Reproduced with permission from [17]. Copyright (1988) American Physical Society.

The fact that the director of LCs, which is often equivalent to the optic axis of the material, is easily influenced by a variety of external stimuli such as mechanical, magnetic, electric, or optic fields, as well as temperature, makes liquid crystals attractive to both the industry and academia [6][18][19][20][21][22][23][24][25][26][27], as exemplified by the \$100 billion industry built around displays and large-screen, flat-panel consumer electronics. Due to the great achievement in displays since the 1970s, LCs are one of the most popular materials around the world [28]. However, with the rapid development and impressive advantages of organic light-emitting displays (OLEDs), there is some competition emerging. Over the last decade, more and more scientists have moved their attention away from display materials to a diversity of new fields—for instance, new optical devices, telecommunication, information storage, energy conservation, elastomer robots, sensors, biotechnologies, nano-/micromanipulation, just to name a few [15][29][30][31][32][33][34][35][36][37][38][39][40][41][42][43][44][45][46]. These new fields can be quite different from displays and more stimulating due to their novelty, leading to a new era for LCs, materials design and technology.

At the same time, revolutionary developments have been made in the fields of nanotechnology and nanoscience, leading to the birth of a series of novel nanomaterials [10][47][48][49][50][51][52][53][54][55]. These nanostructured materials, whose size in at least one dimension is in the range from 1 nm to 100 nm, have gained a wealth of academic and industrial attention due to their size-dependent electronic, optical, magnetic, and chemical properties, which are significantly different from those of bulk materials as well as from individual atoms or molecules [55][56][57][58][59][60][61][62]. These nanostructured materials have been extensively applied in nearly every field of science from energy, optics, computing to catalysis, biosciences and medical sciences [48][63][64][65]. Undoubtedly, when these novel nanometre-scale structures encounter liquid crystals, a highly interesting and unique synergy will be observed, leading to an abundance of entirely new and potential applications [2][8][43][66][67][68][69][70].

The addition of nanomaterials to a LC material produces a composite or colloidal dispersion [71][72]. The new materials are expected to behave differently from their individual components (nanomaterials and LCs) both on the microscopic as well as the macroscopic scale [63][64][65][66][67][68][69][70][71][72][73][74][75][76][77]. There are three basic objectives to making a LC–nanomaterial composite system: (i) to modify the primary physical or chemical properties of the pristine LCs [78][79][80]; (ii) manipulating and ordering nanomaterials in LCs to modify the properties of nanomaterials [81]; and (iii) to obtain additional functionalities that are available from neither the LCs nor the nanomaterials in their intrinsic states [82].

A fundamental investigation of the nanophysics of LCs was published by Brochard and de Gennes, who discussed a theory of magnetic nanoparticles suspended in a nematic LCs in 1970 [83]. Later in the same year, Rault et al. reported the first suspension composed of nematic LCs and small magnetic particles [84]. The main idea of these pioneering studies was to control the director of the LC hosts by a coupling between LC molecules and elongated magnetic particles, producing what has been called magneto-nematics, i.e., nematic liquid crystals with ferromagnetic properties. In fact, it should be mentioned that this conclusion is wrong, as the produced system is not ferromagnetic. As the magnetic field is removed, the magnetization does in fact decrease to zero, while it should retain a finite value for ferromagnetics. Nevertheless, one could call the produced system an anisotropic ferrofluid. Still, this idea led the trend of studies on LC–nanomaterial suspensions for the next few decades. With the rapid development of composite LC materials, it was recognized that long-range orientation and interactions in mesophase can lead to a strong impact of nanomaterials on the properties of the LC hosts [85][86][87] and vice versa; the LC matrix can rearrange the orientational and positional order of nanomaterials [88][89]. The coupling of long-range ordering in LCs and the unique properties of dopant nanomaterials allows us to change or even impose unique physical properties on the LC–nanomaterial composites by doping different kinds of nanomaterials in LCs [70].

Up to now, there have been four primary groups of nanomaterials used as dopants in LCs, i.e., metal nanoparticles, ferroelectric nanoparticles, semi-conductor nanoparticles, and carbon nanoparticles. Among these dispersions, improved physical characteristics and novel functionalities can be obtained depending on the physical and chemical characteristics of the dopants as well as the interaction between LCs and nanomaterials [90][91][92][93][94][95]. For example, there are abundant reports declaring that doping metallic, semiconducting, oxide and ferroelectric nanoparticles into LCs can efficiently modify the electrical and mechanical (viscoelastic) properties of LC host, leading to large dielectric and optical anisotropy, low threshold voltage, and an improved electro-optical response [87][92][94][96][97]. In addition, ferroelectric nanoparticles can also increase the order parameter as well as the clearing point of the LC host due to the interaction between the elastic forces of LC molecules and the spontaneous polarization of ferroelectric nanoparticles [86][98][99][100][101]. Doping nematic LCs with ferromagnetic particles can effectively reorient the director of LCs by magnetic fields induced by the coupling of magnetic particles with LC molecules [83][102][103][104][105][106]. At the same time, there have been reports claiming the exact opposite behaviour: an increase in threshold voltage, a slower optical response, and a decrease in order and in transition temperatures. This indicates that the properties of the composite materials are very much dependent on the dopant size, materials employed, preparation conditions and time. It further indicates that a lot more research is needed to fully understand these systems. With largely varying results, even on a qualitative basis, the field of liquid crystal nanoparticle composites is only in its infancy.

Metal and semiconducting nanoparticles have been used to stabilise the mesophase known as the blue phase, effectively broadening the temperature range of this frustrated phase from 1 K to up to 20 K [107][108]. Silica, ferroelectric and metal nanoparticles can effectively influence the electro-optical response of nematic LCs and induce a memory effect, i.e., a residual transmittance can be maintained without external electric fields being applied [109][110][111][112]. Moreover, a frequency-dependent electro-optical response can be obtained in suspensions of metal nanoparticles and nematic LCs due to the coupling of the dielectric properties of the nanoparticles and the LCs [78][113]. It was also reported that carbon nanotubes dispersed in nematic LCs would align along the direction of the LC director, therefore largely changing the conductivity depending on the orientation of the tubes [114]. In fact, it could be shown that the nanotubes can be reoriented elastically by reorienting the director through applied electric or magnetic fields. Fullerenes could effectively improve the switching speed of a nematic host LC [95], and improved electro-optic responses could be induced by doping graphene oxides in LCs [115].

In addition to changing the intrinsic physical properties of the pristine LC materials mentioned above, intriguing photonic functionalities can also be introduced into the composites by combining nanomaterials with LCs, which is considered a promising method for the building of novel metamaterials [116]. Metamaterials are artificial engineered bulks with regular nanostructures that show exotic optical properties [117]. Combining the emerging field of optical metamaterials with LCs provides an extremely attractive quality, i.e., tunability, which is of the utmost importance in applications such as optically addressed spatial light modulators, tunable photonic materials and dynamic holography [118][119][120]. When integrating LCs with metamaterials, one can modify the director alignment of LCs by applying external stimuli and therefore manipulating the overall optical characteristics of the composite [119]. For instance, upon periodically embedding particular metal nanomaterials, such as gold nanoparticles, into LC matrixes, a localized surface plasmon resonance (LSPR) will be obtained, which can be tuned by adjusting the birefringence of the surrounding LC matrix [77][81][121][122][123][124][125]. Apart from the abovementioned technologies, novel photonic properties and applications can be further exploited for nanodopants including semiconducting nanomaterials, dyes, oxides, quantum dots (QDs), etc. [126][127][128][129]. Instead of the SPR effect in the metallic nanomaterial–LC colloidal systems, the main property of interest in these cases is photoluminescence. The optical excitation and emission of these nanomaterials could be tuned effectively through the interaction between nanomaterials and the long-range ordered LC molecules, leading to a series of potential applications such as information storage, displays, LC lasers, etc. [130][131][132][133][134].

At present, one of the central challenges faced by the development of novel LC–nanomaterial composites is reliable assembly of nanoscale building blocks into functional bulk materials by using distorted LCs. It has already been demonstrated that the localisation of a nanoparticle in a distorted LC region allows the decrease of the free energy, and the directional motion of nanomaterials in LCs can be driven by the collective long-range interactions mediated by the LC director field. As a result, the energetic cost of LC defects enables them to entrap and reorient nanomaterials in a reversible manner and with well-manipulated position and orientation [135][136][137][138]. In addition, nanomaterials that are suspended in LCs and have a given anchoring energy at the surface will induce a deformation of the orientation in the surrounding LCs, producing topological defects in the vicinity of the nanoparticle. The elastic distortion exerts a force on neighbouring nanomaterials at a range of up to a few micrometres, resulting in defect lines or points around nanomaterials [139][140][141][142][143][144]. This LC-mediated interaction can be either repulsive or attractive and, as a consequence, can be used as an effective method to manipulate the spatial distance between nanoparticles [88][145][146][147][148]. It is thus anticipated that, by coupling these two LC-mediated effects will result in the assembly of nanomaterials with one-, two- or

even three-dimensional (3D) periodic superstructures <sup>[143][149][150][151][152][153][154]</sup>. Due to the great potential in both fundamental science and applications, this top-down assembly approach, where spontaneous or artificially generated LC textures are utilized as templates for organizing nanoparticles on controlled lattices or confining them in designed defects, has been increasingly coming into the spotlight.

Another interesting research field of LC–nanomaterial technology that is worthwhile to discuss is the self-assembly of lyotropic LCs composed of 1- or 2D anisotropic nanomaterials <sup>[3][7]</sup>. This topic has been investigated from time to time as people have realized that suspensions of anisodiametric nanomaterials can form LC phases, even at a very low concentration, due to Bernal's seminal work on suspensions of tobacco mosaic virus <sup>[155]</sup> and Onsager's theory of the excluded volume mechanism <sup>[156]</sup>. However, with the boom in the development of nanotechnology and nanoscience in the last decade, various types of novel nanomaterials have emerged. The size dispersion, shape anisotropy, and surface morphology of the nanomaterials are now well controlled. All of these developments will definitely lead to a dramatic increase in the use of nanoparticle-based LCs. Nowadays, it is well known that suspensions of anisotropic nanorods or nanoplates, viruses, nano-cellulose, carbon nanotubes and graphene oxide can form lyotropic LC phases, which are usually accompanied by surprising physical properties <sup>[157][158][159][160][161][162][163][164][165]</sup>.

## **2. Summary**

Here, we summarized the influence of nanoparticle doping on the electro-optical and other physical properties, of liquid crystals, including molecule alignment, viscosity, clearing point, or elastic constants, just to name a few of the properties introduced. Various kinds of nanodopants, such as ferroelectric nanoparticles, noble metallic nanoparticles, semiconductor nanoparticles and carbon nanoparticles, have been presented to induce modifications of the physical properties of LCs. However, these studies create an inconsistent picture of the beneficial effects of nanoparticles on the physical properties of LC hosts. This may be attributed to the effect of different nanoparticles' size, shape, dispersibility and functionalization. To thoroughly understand the influence of nanoparticle doping on the physical properties of LCs, further systematic investigations are required. We further demonstrated the exciting photonic functionalities, including SPR effect, photothermal effect and lasing, of LC–nanoparticle composites. The intrinsic optical properties of nanoparticles can be effectively modified by a LC medium, leading to a variety of multifunctional photonic applications.

Various nanoparticle mesostructures, obtained either within distorted areas of LCs or topological defects, were discussed. The self-organization-assisted nanoparticle assembly within the disclination lines will effectively eliminate high-energy volume areas and reduce the elastic-free energy density of the LC system. Such an effect can even lead to a stabilization of the 3D blue phase defect lattice.

Additionally, we demonstrated the self-assembly of nanoparticles into lyotropic LC phases. Colloidal suspensions of anisotropic particles, including rods, tubes, disks, flexible chains and wires, can self-organize from the isotropic disordered to the nematic ordered phase as the concentration exceeds a critical value according to Onsager's model. These lyotropic LC materials have intriguing physical properties and can be applied to various functional devices.

Finally, we briefly introduce the practical applications of LC–nanoparticle composites in biological systems. The aspects of biological sensors and drug delivery systems are of particular interest, and it is anticipated that this area of novel liquid crystal research will attract much interest in the liquid crystal community and soft matter community in particular.

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