TOPICAL REVIEW

Conventional and unconventional ionic phenomena in tunable soft materials made of liquid crystals and nanoparticles

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Abstract
A great variety of tunable multifunctional materials can be produced by combining nanoparticles and liquid crystals. Typically, the tunability of such soft nanocomposites is achieved via external electric fields resulting in the field–induced reorientation of liquid crystals. This reorientation can be altered by ions normally present in liquid crystals in small quantities. In addition, nanomaterials dispersed in liquid crystals can also affect the behavior of ions. Therefore, an understanding of ionic phenomena in liquid crystals doped with nanoparticles is essential for future advances in liquid crystal-aided nanoscience and nanotechnology. This paper provides an overview of the ionic effects observed in liquid crystals doped with nanomaterials. An introduction to liquid crystals is followed by a brief overview of nanomaterials in liquid crystals. After giving a basic description of ions in liquid crystals and experimental methods to measure them, a wide range of ionic phenomena in liquid crystals doped with different types of nanomaterials is discussed. After that, both existing and emerging applications of tunable soft materials made of liquid crystals and nanodopants are presented with an emphasis on the role of ionic effects in such systems. Finally, the discussion of unsolved problems and future research directions completes the review.

Introduction

Liquid crystals (LC) are a thermodynamically stable state of matter that occurs between a crystalline solid and isotropic liquid [1–3]. Liquid crystals combine the properties of liquids and crystals. They can flow like a liquid, and also exhibit crystal-like behaviour (i.e. an anisotropy of their physical properties) because of a long-range orientational order of anisometric molecules making up liquid crystals [1–3]. Existing liquid crystal materials can be broadly categorized in three basic groups, namely thermotropic liquid crystals, lyotropic liquid crystals, and liquid crystal polymers as shown in figure 1(A). In the case of thermotropic liquid crystals, the mesomorphic (i.e. liquid crystalline) phase is achieved by varying the pressure and/or temperature. A series of phase transitions involving thermotropic liquid crystal (smectic and nematic) phases is shown in figure 1(B). Lyotropic phases are obtained by changing the concentration of a dopant in a solvent. Low molecular weight thermotropic liquid crystals are made of anisometric molecules whereas lyotropic liquid crystals are typically formed via the self-assembly of amphiphilic molecules, dyes, clays, nanoobjects, and viruses [1]. Polymer liquid crystals are made of macromolecules characterized by high molecular weight [1].

Thermotropic liquid crystals can be further categorized into molecular liquid crystals, ionic liquid crystals, and liquid crystalline semiconductors. The present review focuses on molecular liquid crystals. A few words should be mentioned about the difference between ionic liquid crystals and molecular liquid crystals. Ionic liquid crystals behave as anisotropic ionic conductors. They exhibit intrinsic ionic conductivity $\lambda_{DC}$ due to their chemical composition ($\lambda_{DC}$ can be as high as $1$–$10$ S m$^{-1}$). Existing and emerging applications of ionic liquid crystals are centered around this basic physical property [4]. On the contrary, molecular liquid crystals are anisotropic dielectric materials. A finite electrical conductivity of molecular liquid crystals (normally, in the $10^{-7}$–$10^{-12}$ S m$^{-1}$ range) is caused by ionic impurities added to a mesogenic host intentionally or
unintentionally [3]. A wide range of applications of molecular liquid crystals utilize orientational transitions of a liquid crystal director caused by external fields. Ionic liquid crystals and liquid crystalline semiconductors are covered in several reviews and books [4–6]. Nanostructured liquid crystal electrolytes and conductors [7] are also beyond the scope of this paper.

Molecular liquid crystals exhibit an anisotropy of their physical properties. This anisotropy can be tuned by external factors such as electric and magnetic fields. Such fields can control the average orientation of mesogenic molecules making up liquid crystals thus tuning their physical properties (refractive indices, dielectric constants, etc.). This fact has resulted in numerous applications of liquid crystals. They include different types of liquid crystal displays (LCD), both conventional displays [8] and advanced displays for virtual and augmented reality [9], tunable optical components (lenses, filters for hyperspectral imaging [10, 11], waveplates and retarders [11, 12], diffractive optical elements [13]), liquid crystal on silicon (LCoS) devices [14], spatial light modulators and beam-steering devices [14], light shutters [15, 16], smart windows [17–19], reconfigurable signal processing devices operating at microwave and millimeter wave frequencies [20, 21], and countless others. A sensitivity of liquid crystals to various stimuli has also been employed in the design of chemical and biological sensors [22].
Future progress in liquid crystal science and liquid crystal technology relies on the development of advanced liquid crystal materials. A great variety of tunable multifunctional materials can be produced by combining nanoparticles and liquid crystals.

**Nanomaterials in liquid crystals**

Nanomaterials in liquid crystals are a hot topic of contemporary liquid crystal research. The number of research papers, special issues, books, and reviews published on the subject of merging nanoscience and liquid crystals unambiguously support this statement. The idea to modify the properties of liquid crystals by mixing them with magnetic fine particles (i.e. nanoparticles) (expressed by Brochard and de Gennes back in the 1970s [23]) became increasingly popular since the rise of nanotechnology in the 1990s. Since that time a great variety of nanomaterials in liquid crystals have been studied by independent research groups (figure 2). There are several reasons for ongoing and very active research activities in this field.

By adding nanomaterials to liquid crystals new composite materials can be created. They can exhibit the combined functionality of liquid crystals and nanodopants, often unachievable by means of chemical synthesis. For example, such exotic materials as ferromagnetic nematic liquid crystals can become a reality [24]. Very often, the combination of liquid crystals and nanomaterials can result in quite unusual behavior of such systems [25]. In general, nanomaterials mixed with liquid crystals modify their physical and chemical properties. Liquid crystals can also be used to order nanomaterials and control their self-assembly and serve as nanoreactors for template synthesis of nanomaterials [26, 27]. Given a very large number of research papers published in this field it is impossible to give a comprehensive overview of all effects observed in liquid crystals doped with nanomaterials. An interested reader is referred to numerous reviews on nanomaterials in liquid crystals discussing in greater detail various aspects of such systems. A brief overview of review papers on nanomaterials in liquid crystals is given in this section.

There are several reviews covering major types of nanomaterials in different types of liquid crystals. A series of papers by Hegmann et al published in the early 2010s provide a comprehensive overview of nanoparticles in liquid crystals including their classification, chemical synthesis, self-assembly, defect formation, new functionalities, and potential applications [28–30]. Nanomaterials of different types in various liquid crystal systems were further discussed by Garbovskiy and Glushchenko [31]. Self-assembly and ordering of nanoparticles in liquid crystals were analyzed by Bisoi and Kumar [32], Blanc et al [33], and Saliba et al [34]. The breadth of possibilities offered by the combination of liquid crystals and nanomaterials was comprehensively discussed in several collective monographs edited by Li [35], Lagerwall and Scalia [36], and Dierking [37]. An overview of recent progress, current trends, and future research directions in this highly multidisciplinary field combining liquid crystals, nanoscience, and nanotechnology can be found in nicely written papers just appeared in print [38–40].

A constant growth and expansion of research into the properties of nanomaterials in liquid crystals resulted in a rapid increase in the number of published topical reviews with an emphasis on a particular class of nanodopants in liquid crystals. An overview of science and technology of carbon nanotubes in liquid crystals was given by Lagerwall and Scalia [41], Rahman and Lee [42], Dolgov et al [43], Yadav and Singh [44]. Liquid crystals can control the ordering of carbon nanotubes (CNT) whereas CNT can modify the properties of liquid crystals resulting in faster electro-optical response, enhanced dielectric constant and altered resistivity, large nonlinear-optical and memory effects, just to name a few (references to original publications can be found in reviews [41–44]). Moreover, under certain conditions carbon nanotubes and other carbon based nanomaterials such as graphene oxides can exhibit liquid crystal phases [45–47] and can be considered as potential materials for the design of ITO-free electrodes (interested readers are referred to recent comprehensive review by Dierking [48]).

Semiconductor nanomaterials, especially quantum dots and nanorods in liquid crystals, are receiving increasing attention due to their very intriguing properties suitable for the design of tunable light emissive composite materials. Liquid crystals can tune the spectrum, polarization, and isotropy of the emission while semiconductor quantum scale particles can alter physical properties of liquid crystals such as viscoelastic constants, dielectric permittivity, electrical conductivity, and switching parameters. In addition, liquid crystals can be used to manipulate and control the self-assembly of semiconductor nanoparticles. Several review papers by Mirzaei et al [49], Singh et al [50, 51], and Prevot et al [52] provide a detailed overview of recent progress in this vibrant field of research.

The tunability of liquid crystals offers an exciting opportunity to design reconfigurable plasmonic materials made of plasmonic nanoparticles and a mesogenic host. An active control over the frequency of the plasmonic resonance of metal nanoparticles dispersed in anisotropic fluids can be achieved by taking advantage of the sensitivity of liquid crystals to external stimuli. Different aspects of liquid crystals doped with metal nanoparticles, composite materials enabling liquid crystal based plasmonics, were discussed in a series of review...
papers by Qi and Hegmann [53], Abdulhalim [54], Si et al [55], Lewandowski et al [56], Choudhari et al [57], Atorf et al [58], and Chen et al [59].

Seminal paper by F Brochard and P G de Gennes initiated a very active research into the properties of magnetic nanoparticles in liquid crystals. Magnetic nanomaterials can increase the sensitivity of liquid crystals to magnetic fields and make possible the control over orientational transitions in liquid crystals. Recent advances in this rapidly growing field are presented in comprehensive reviews by Tomasovicova et al [60], Mertelj and Lisjak [61], and Hähsler et al [62].

Ferroelectric nanoparticles respond to electric fields, and, if mixed with liquid crystals, can increase the sensitivity of mesogenic materials to electric fields. This line of research was initiated by Reznikov in the early 2000s. Since that time numerous papers revealed a variety of intriguing effects of ferroelectric nanoparticles on the properties of liquid crystals summarized in several reviews [63–65]. Photorefractive applications of liquid crystals doped with ferroelectric nanoparticles were discussed by Evans et al [66].

Important parameters of liquid crystal materials such as the threshold voltage, viscoelastic constants, birefringence and dielectric anisotropy can be modified in a desirable way by mixing them with metal oxide nanoparticles. An overview of basic properties and potential applications of metal oxide nanoparticles in liquid crystals was given in a paper by Prakash et al [67].

Figure 3. (A) Typical electro-optical experiment utilizing nematic liquid crystals placed between crossed polarizers. (B) Screening effect caused by ions in molecular liquid crystal.
There are also topical reviews discussing different effects of nanomaterial on electro-optical properties of liquid crystals [68, 69], on the properties of ferroelectric liquid crystals [70], on the stability and phase formation of liquid crystals [71], on the dynamics of dislocations in smectic A liquid crystals [72], and on the formation of lattices of topological defects [73]. The properties of discotic liquid crystals doped with nanomaterials are summarized in reviews by Kumar and coworkers [74–76]. Finally, several review papers discuss biological aspects [77] and biomedical applications [78–82] enabled by the combination of nanotechnology and liquid crystals.

Review papers we just discussed unequivocally indicate a wide range of applications enabled by tunable composite materials made of thermostropic liquid crystals and nanoparticles. Typically, the tunability of such soft nanocomposites is controlled by external electric fields resulting in the field-induced reorientation of liquid crystals. This reorientation can be altered by ions normally present in liquid crystals in small quantities (in the simplest case, through a well-known screening effect, this effect will be explained in greater detail in the next section). In addition, nanomaterials dispersed in liquid crystals can also affect the behavior of ions. Therefore, an understanding of ionic phenomena in liquid crystals doped with nanoparticles is essential for future advances in liquid crystal-aided nanoscience and nanotechnology. An overview of ionic effects observed in liquid crystals doped with nanomaterials can be found in paper by Garbovskiy and Glushchenko [83]. Reference [83] provides an analysis of papers reporting ionic effects in liquid crystals/nanoparticles systems published prior to 2015. In this topical review, recent advances in ionic phenomena observed in liquid crystals doped with nanomaterials are discussed. An emphasis is placed on progress that has been made since 2015.

**Ions in liquid crystals and how to measure them**

Prior to discussing sources of ions in molecular liquid crystals, it is instructive to show how ions can actually alter the performance of liquid crystal devices. Consider a standard electro-optical effect employed in many liquid crystal devices and shown in figure 3. A sandwich-like cell filled with homogeneously aligned nematic liquid crystals is placed between two crossed polarizers. The angle between the liquid crystal director and the axis of polarizers is ±45°, respectively. An intensity of light passing through the cell is measured as a function of voltage applied across the cell (figure 3(A)). If the applied voltage is greater than the threshold voltage, a liquid crystal director will be reoriented along the field (assuming a positive dielectric anisotropy of nematics). The degree of the reorientation depends on the value of the applied electric field. The reorientation of liquid crystals leads to a change in the effective birefringence $\Delta n$. Because of this change, the intensity of the light passing through the cell exhibits an oscillatory dependence according to equation (1):

$$I = I_0 \sin^2(2\beta) \sin^2\left(\frac{\pi d \Delta n}{\lambda}\right)$$

where $I_0$ is the intensity of the incident light, $\beta$ is the angle between the liquid crystal director (i.e. an optical axis of uniaxial nematic liquid crystals) and the polarization of light of the incident light and (in the case shown in figure 3, $\beta = 45^\circ$), $d$ is the cell thickness, $\Delta n$ is the birefringence of liquid crystals, and $\lambda$ is the wavelength of the laser beam (in the case shown in figure 3, $\lambda = 632.8$ nm) [1]. Depending on the phase shift $\Delta \Phi = \frac{2\pi d \Delta n}{\lambda}$, several peaks can be observed (one peak corresponds to a $2\pi$ change in the phase shift). Equation (1) can be used to compute the phase change using experimentally measured normalized transmittance of the liquid crystal cell defined by the ratio $I/I_0$. Figure 3(A) shows that both the amplitude and the phase of light can be controlled by employing orientational transitions in liquid crystals. This simple idea can be applied to a great variety of applications.

Ions present in molecular liquid crystals can alter the orientational transitions in liquid crystals through a screening effect as shown in figure 3(B). The applied voltage reorients the liquid crystal director towards the normal to the substrates of the cell (the direction of the electric field). This reorientation results in a measured normalized transmittance (a part of the curve marked with number 1, figure 3(B)). The achieved nearly $2\pi$ change in the phase shift corresponds to the point A on the measured curve (figure 3(B)). Ions in liquid crystals alter this reorientation and the achieved change in the phase shift in the following way. The applied electric field separates positive and negative ions as shown in figure 3(B) (positive ions move to the negatively charged electrode, and negative ions move in the direction of the positively charged electrode). Those separated ions create their own electric field $E_{\text{ions}}$ (dashed arrow, figure 3(B)) acting against an external electric field $\vec{E}$ (solid arrow, figure 3(B)). If the magnitude of the combined electric field $(\vec{E} - E_{\text{ions}})$ becomes smaller than the threshold value, the liquid crystal director will be reoriented back to its initial state (a planar alignment) by restoring elastic forces. This reorientation of liquid crystals back to their initial planar alignment caused by the screening effect corresponds to a part of the measured transmittance marked with number 2, figure 3(B). As a result, instead of the expected nearly $2\pi$ change in the phase shift we obtained no change at all, even though the electric field was still applied across the cell.
Figure 3 shows that ions can alter the performance of liquid crystal devices. In the case of displays, ions in liquid crystals can lead to an image sticking, image flickering, reduced voltage holding ratio, and overall slow response thus affecting an image quality and switching \[84, 85\]. A better understanding of sources of ions in liquid crystals and mechanisms of their generation could help to reduce or even eliminate the aforementioned negative effects of ions in liquid crystals. That’s why ions in molecular liquid crystals have been studied since the early 1960s. According to existing literature, ions can be generated both in the bulk of liquid crystals and on the surface of the liquid crystal cell \[3, 85, 86\]. More specifically, ions in liquid crystals can be generated due to the dissociation of neutral molecules \[87\]. The ionic contamination through chemical synthesis (chemical leftovers) is also an important source of ions \[88, 89\]. The decomposition of liquid crystal materials because of aging is also a very common source of ions \[90\]. In addition, ions can be generated by ionizing radiation such as UV light \[91–93\], and gamma-radiation \[94, 95\], by high electric fields \[96–98\], and by means of electro-chemical reactions \[99\]. Ions can be introduced intentionally by doping them with ionic species \[100, 101\]. They can also be generated in the glue used to seal the liquid crystal cell \[102\]. Contaminated alignment layers are also a very common source of ions in liquid crystals \[103–106\]. Recently, a new source of ions, namely, nanomaterials in liquid crystals, has been reported \[83, 107, 108\]. It will be discussed in more detail in the next sections of this review.

Information about ions in liquid crystals can be obtained by performing electrical measurements. Ions result in a DC electrical conductivity of liquid crystals \(\lambda_{DC}\) written as (2):

\[
\lambda_{DC} = \sum_i q_i \mu_i n_i
\]

where \(n_i\) is a volume concentration of the \(i\)th ions (ion density), \(q_i\) is a charge of the \(i\)th ion, and \(\mu_i\) is their mobility. In the simplest case of two monovalent ions, the electrical conductivity can be rewritten as (3)

\[
\lambda_{DC} = |e|(\mu^+ + \mu^-)n = |e| \mu n
\]

where \(|e| = 1.6 \times 10^{-19}\text{C}\), and \(\mu = \mu^+ + \mu^-\) (the effective mobility).

Physical quantities of interest (the electrical conductivity, the ion mobility, and the concentration of ions) can be obtained by applying methods of transient current measurements \[86, 109–111\] and dielectric spectroscopy \[112–115\]. The combination of electrical and electro-optical measurements can also be utilized \[116\]. Figure 4 provides a summary of typical experimental techniques and steps involved in measuring ions in liquid crystals.

Measurements of an AC electrical conductivity is a relatively straightforward process, whereas an evaluation of a DC conductivity, ion mobility, and ion density is a challenging task \[112, 115, 117\]. A standard liquid crystal cell has a multilayer structure. Each glass substrate of a sandwich-like cell is covered with an electrically conductive ITO (indium tin oxide) layer with a thin alignment layer on a top of an ITO layer. A thin layer of liquid crystals is sandwiched between two such substrates. As a result, a suitable model is needed to extract the DC conductivity of liquid crystals \(\lambda_{DC}\) using the measured dielectric spectra \(\varepsilon = \varepsilon' - i\varepsilon''\) or complex electrical conductivity \(\lambda = \lambda' + i\lambda''\). Quantities \(\varepsilon\) and \(\lambda\) are related according to equation (4):
where \( f \) is a frequency, and \( \varepsilon_0 = 8.854 \times 10^{-12} \text{ F m}^{-1} \). The discussion of different ways to evaluate the DC electrical conductivity of liquid crystals can be found in recent paper by Karaawi et al [115]. The obtained values of DC conductivity can be used to evaluate the concentration of ions if their mobility is known.

The measured impedance spectra \( Z = Z' - iZ'' \) can also be used to estimate the concentration of ions and their diffusion coefficients \( D (D' \mu) \) by fitting an appropriate model to the measured data as explained in publications by Sawada et al [114], Barbero et al [113], and Khazimullin et al [112]. It should be mentioned that an interpretation of measured dielectric spectra is always a topic of hot discussions [118–120].

Transient current measurements provide valuable information about the concentration of ions in liquid crystals [86, 109–111]. The measured transient current \( i(t) \) can be expressed as a sum of a capacitive current \( i_{\text{cap}} \), conduction current \( i_c \), and polarization switching current \( i_p \) (to account for possible ferroelectric response), according to (5):
where $t$ is time. The concentration of ions can be estimated according to

$$i(t) = i_{\text{cap}}(t) + i_i(t) + i_p(t)$$  \hspace{1cm} (5)

where $d$ is a cell thickness, $A$ is the area of electrodes, and $T/2$ is the integration interval.

Transient current measurement can also be used to evaluate the effective mobility of ions by observing an ‘ion bump’ (a maximum of current) in a liquid crystal cell driven by low frequency square waves with different DC offsets. The effective mobility is computed according to (7):

$$\mu = \frac{d^2}{V t_{\text{trans}}}$$  \hspace{1cm} (7)

where $t_{\text{trans}}$ is the time position of the observed maximum of current, and $V$ is the magnitude of the applied voltage [86, 109–111].

It should be noted that both electrical conductivity and ion mobility of liquid crystals are anisotropic quantities. The quality of alignment of liquid crystal nanocomposites should be carefully checked and adjusted during experiments in order to measure all components of the electrical conductivity tensor. Measurements of basic parameters of ions in liquid crystals is a very delicate process, and improvements are needed in both experimental techniques and our interpretation of the measured data [112, 115, 117]. This important aspect will be discussed later in the concluding sections of this review.

### Ionic effects in liquid crystals doped with nanomaterials

We already emphasised in earlier sections that the dispersion of nanomaterials in liquid crystals has emerged as a promising way to modify their properties and design novel materials suitable for many applications. From the perspective of ion generation in liquid crystals, a very important question is, how can nanodopants affect the behaviour of ions in liquid crystals. Ion-related effects of nano-objects in liquid crystals were reported in many publications. Papers published prior to 2015 have been reviewed by Garbovskiy and Glushchenko (2016) [83]. In this review, papers published since 2015 will be analyzed with an emphasis on recent advances in our understanding of ion phenomena in liquid crystals doped with nanomaterials.

An analysis of research papers performed back in 2015 revealed the existence of several regimes observed in liquid crystals doped with nanomaterials. A great variety of existing experimental results on ions and nano-objects in liquid crystals can be broadly categorized into the following groups: (i) publications presenting a decrease in the concentration of ions in liquid crystals (the ion trapping regime); (ii) studies reporting an increase in the concentration of ions (the ion generation regime); (iii) the combination of both ion trapping and ion generation regimes (depending on the concentration of nanodopants in liquid crystals) [83] and references therein. A very important result is that the same type of nanomaterials dispersed in different liquid crystals can lead to different regimes (ion trapping or ion generation) [121, 122]. An elementary quantitative model of these regimes was proposed in 2016 by Garbovskiy [123, 124]. This model introduced the ionic contamination of nanomaterials as a key factor enabling the possibility of different regimes (ion trapping or ion capturing regime), ion generation (or ion releasing regime), and no change regime in liquid crystals doped with nanomaterials [107, 108, 124] (figure 5).

If nanodopants are 100% pure, the ion trapping regime is the only possible outcome of their dispersion in liquid crystals (figure 5(C), dashed-dotted curve). On the contrary, liquid crystals doped with contaminated nanoparticles can exhibit three different regimes, namely, the ion trapping regime (figure 5(C), solid curve), ion generation regime (figure 5(C), dashed curve), and no change regime (figure 5(C), dotted curve). Both ion trapping and ion generation regimes are characterized by the saturation effect (figure 5(C), solid and dashed curves). Switching between these regimes is governed by the interplay between the ionic purity of nanoparticles,
the ionic purity of liquid crystals, and the ratio of the ion releasing and ion capturing rate constants (constant $K_{NP}$) as shown in table 1.

The proposed approach, within the limits of its applicability discussed in papers [123, 125], can describe a broad variety of existing, even seemingly contradictory, experimental data reported for ferroelectric nanoparticles in liquid crystals (table 2), carbon-based nanoobjects in liquid crystals (table 3), and semiconductor and dielectric nanodopants in liquid crystals (table 4).

Selected case studies listed in table 4 (experimental results obtained by Shcherbinin and Konshina [133, 134], and by Kovalchuk et al. [135]) are shown in figure 6. As can be seen from figure 6, the reported experimental data [133–135] and the model [108] are in very good agreement. The proposed model can also be generalized to account for several types of ionic species in liquid crystals [123, 137, 138], temperature-induced effects [129, 139], and the presence of alignment layers [138, 140, 141].

The results summarized in tables 2–4 and figure 6 unambiguously indicate that nanomaterials in liquid crystals can behave as a source of ions or as ion traps. At the same time, they do not cover an entire spectrum of new research findings appeared in print since 2015. It is both interesting and instructive to provide a brief overview of such publications.

| Table 2. Case studies: Reported experimental data on ferroelectric nanoparticles in liquid crystals, and physical parameters of the model proposed in [123]. |
| --- |
| Materials | Reported effects | Physical parameters |
| Ferroelectric nanoparticles (LiNbO₃) in liquid crystals | Ion capturing effect [126] | $K_{NP} = 7 \times 10^{-21} \text{ m}^3$; $\nu_{NP} = 0.1075$; $\sigma_{NP}^s = 5 \times 10^{18} \text{ m}^{-2}$; $R_{NP} = 12.5 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 4.65$ [124] |
| Ferroelectric nanoparticles (BaTiO₃) in nematic liquid crystals | Ion capturing effect [127] | $K_{NP} = 4 \times 10^{-26} \text{ m}^3$; $\nu_{NP} = 0.3$; $\sigma_{NP}^s = 10^{19} \text{ m}^{-2}$; $R_{NP} = 1000 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 6.02$ [124] |
| Ferroelectric nanoparticles (BaTiO₃) in nematic liquid crystals (E44) | Temperature-induced release of ions [128] | $\nu_{NP} = 0$; $K_{NP}^s = 1.93 \times 10^{-33} \text{ m}^3$; $\Delta E = 0.4 \text{ eV}$; $\sigma_{NP}^s = 5 \times 10^{18} \text{ m}^{-2}$; $R_{NP} = 20 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 6.02$ [129] |

| Table 3. Case studies: Reported experimental data on carbon-based nanoobjects in liquid crystals and physical parameters of the model proposed in [123]. |
| --- |
| Materials | Reported effects | Physical parameters |
| Carbon nanotubes (CNT) in nematic liquid crystals (E7) | Ion capturing effect [130] | $K_{NP} = 0.7 \times 10^{-21} \text{ m}^3$; $\nu_{NP} = 9.5 \times 10^{-4}$; $\sigma_{NP}^s = 10^{19} \text{ m}^{-2}$; $R_{CNT} = 2.5 \text{ nm}$; $L_{CNT} = 500 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 1.61$ [124] |
| Diamond nanoparticles in nematic liquid crystals (E7) | Ion capturing effect [121] | $K_{NP} = 10^{-22} \text{ m}^3$; $\nu_{NP} = 10^{-2}$; $\sigma_{NP}^s = 1.25 \times 10^{17} \text{ m}^{-2}$; $R_{NP} = 5 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 3.3$ [124] |
| Diamond nanoparticles in nematic liquid crystals (E7) | Ion releasing effect [121] | $K_{NP} = 0.8 \times 10^{-23} \text{ m}^3$; $\nu_{NP} = 0.25$; $\sigma_{NP}^s = 1.25 \times 10^{17} \text{ m}^{-2}$; $R_{NP} = 5 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 3.3$ [124] |
| Graphene nano-flakes (GNF) in nematic liquid crystals (8OCB) | Ion capturing effect [131] | $K_{GNF} = 0.8 \times 10^{-23} \text{ m}^3$; $\nu_{GNF} = 8.5 \times 10^{-3}$; $\sigma_{GNF}^s = 0.33 \times 10^{18} \text{ m}^{-2}$; $R_{GNF} = 5 \text{ nm}$; $L_{GNF} = 10 \text{ nm}$; $\rho_{GNF}/\rho_{LC} = 1.8$ [124] |

| Table 4. Case studies: Reported experimental data on dielectric and semiconductor nanomaterials in liquid crystals and physical parameters of the model proposed in [123]. |
| --- |
| Materials | Reported effects | Physical parameters |
| Anatase (TiO₂) nanoparticles in nematic liquid crystals (E44) | Ion capturing effect [132] | $K_{NP} = 10^{-21} \text{ m}^3$; $\nu_{NP} = 1.5 \times 10^{-4}$; $\sigma_{NP}^s = 0.8 \times 10^{18} \text{ m}^{-2}$; $R_{NP} = 5 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 3.9$ [124] |
| TiO₂ nanoparticles in nematic liquid crystals (ZhK1282) | Ion releasing effect [133] | $\nu_{NP} = 4.35 \times 10^{-4}$; $K_{NP} = 1.6 \times 10^{-23} \text{ m}^3$; $\sigma_{NP}^s = 0.8 \times 10^{18} \text{ m}^{-2}$; $R_{NP} = 25 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 3.9$ [108] |
| TiO₂ nanoparticles in nematic liquid crystals (ZhK1282) | Ion capturing effect [133] | $\nu_{NP} = 0$; $K_{NP} = 3.65 \times 10^{-24} \text{ m}^3$; $\sigma_{NP}^s = 2 \times 10^{18} \text{ m}^{-2}$; $R_{NP} = 25 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 3.9$ [108] |
| CdSe/ZnS core/shell nanoparticles in nematic liquid crystals (ZhK1289) | Ion releasing effect [134] | $\nu_{NP} = 3.379 \times 10^{-3}$; $K_{NP} = 10^{-26} \text{ m}^3$; $\sigma_{NP}^s = 10^{18} \text{ m}^{-2}$; $R_{NP} = 3 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 5.091$ [108] |
| CuPS₃ nanoparticles in nematic liquid crystals (6CB) | Ion releasing effect [135] | $\nu_{NP} = 0.3075$; $K_{NP} = 10^{-23} \text{ m}^3$; $\sigma_{NP}^s = 7 \times 10^{18} \text{ m}^{-2}$; $R_{NP} = 58.5 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 4.907$ [108] |
| Silica nanoparticles in nematic liquid crystals (E7) | Both ion releasing and ion capturing [136] | $\nu_{NP} = 0$; $K_{NP} = 1.1 \times 10^{-23} \text{ m}^3$; $\sigma_{NP}^s = 2.5 \times 10^{18} \text{ m}^{-2}$; $\nu_{NP} = 2 \times 10^{-4}$; $K_{NP} = 3 \times 10^{-28} \text{ m}^3$; $n_{NP} = 5.39 \times 10^{10} \text{ m}^{-3}$; $\sigma_{NP}^s = 2.5 \times 10^{18} \text{ m}^{-2}$; $n_{NP} = 0 \text{ m}^{-3}$; $R_{NP} = 3.5 \text{ nm}$; $\rho_{NP}/\rho_{LC} = 2.4$ [107] |
Current research into the electrical properties of nanomaterials in liquid crystals is characterized by increased breadth and depth. Independent research groups continue to study electrical properties of thermotropic liquid crystals doped with nanomaterials gradually expanding classes of mesogenic materials and nanodopants. As a rule, basic electrical parameters of liquid crystals are measured as a part of material characterization of liquid crystals doped with nanoparticles. The measured properties are very sensitive to numerous factors. As a result, the reported results may vary, very often, in a substantial way. This variability reflects the complexity of systems under test.

Interesting results were obtained by Urbanski and Lagerwall [142, 143]. They showed that functionalized gold nanoparticles can enrich nematic liquid crystals with ions in a very efficient way. Ionic contaminants trapped by the ligands used to functionalize gold nanoparticles were released in the liquid crystal bulk [142, 143]. Measurements of DC electrical conductivity and improvements in electro-optics of ferroelectric liquid crystals doped with gold nanospheres were reported by Podgornov et al [144, 145]. Gold nanoparticles of different shape were functionalized with alkyl acrylate polymer [144]. The observed faster electro-optical response was attributed to the ion capturing by gold nanospheres [145]. Improvements in electro-optics of nematic liquid crystals doped with gold nanoparticles were recently observed by Chausov et al [146]. A shorter response time, smaller threshold voltage, and an increase in the optical and dielectric anisotropy were associated with the ion capturing effect by gold nanoparticles [146].

A tunability and enhancement of photoluminescence along with the ion trapping effect in cholesteric liquid crystals doped with functionalized silver nanoparticles were reported by Middha et al [147]. Ion trapping by functionalized silver nanoparticles in liquid crystals resulting in better electro-optical performance was also reported by Shivaraja et al [148, 149] for nematic, and by Debnath et al for ferroelectric liquid crystal materials [150]. Silver nanoparticles added to alignment layers of liquid crystal cells can also capture ions and reduce residual direct current voltage as was shown by Liu et al [151]. An increase in the AC electrical conductivity caused by silver nanoparticles in liquid crystals was observed by Katiyar et al [152].

Colloids of dielectric and semiconductor nanomaterials in liquid crystals have also received increasing attention in recent years. Chandran et al [153] carried out electrical and electro-optical experiments demonstrating the ion capturing effect resulting in the non-volatile memory behaviour of ferroelectric liquid crystals doped with nickel oxide nanorods. The electrical behaviour of tin dioxide nanoparticles in liquid crystals was studied by several research groups [133, 154–159]. Both increase and decrease in the concentration of ions in nematic liquid crystals doped with tin oxide nanoparticles was reported by Konshina and her research group.

Figure 6. (A) and (B) The concentration of ions in liquid crystals doped with TiO₂ nanoparticles as a function of the weight concentration of nanoparticles. Experimental datapoints are from [133]. (C) The concentration of ions in liquid crystals doped with Cu₇PS₆ nanoparticles as a function of the weight concentration Experimental datapoints are from [135]. (D) The concentration of 'slow' ions in liquid crystals doped with CdSe/ZnS quantum dots as a function of the weight concentration of nanodopants. Experimental datapoints are from [134]. Reproduced from [108], under the Creative Commons Attribution License.
Yadav al [155] studied effects of ion trapping behavior of TiO2 nanoparticles on physical parameters of weakly polar nematic liquid crystal, while Agrahari et al [156] observed a three order of magnitude reduction of the ion density in ferroelectric liquid crystal mixture doped with tin oxide nanoparticles as compared to that of the undoped liquid crystal mixture [156]. An increase in electrical conductivity and concentration of ions in bent core liquid crystals as a result of mixing them with tin oxide nanoparticles was reported by Nafees et al [157]. It was also found that tin oxide nanoparticles added to nematic liquid crystals can decrease an impedance (Marzal et al [158]) and increase the amplitude of transient currents in such systems (Ayebo et al [159]) suggesting the possibility of the increase in the concentration of mobile ions in such systems.

The ion trapping phenomenon was considered a possible reason for a memory effect in ferroelectric liquid crystals doped with zinc oxide nanoparticles observed by Chaudhary et al [160]. Interestingly, zinc oxide nanoparticles added to nematic liquid crystals resulted in an increase in the contrast ratio, birefringence, and alternating-current conductivity of the dispersion as was reported by Sharma et al [161].

Silica (silicon dioxide) nanoparticles mixed with liquid crystals can result in several ionic effects [162, 163]. A nearly one order of magnitude decrease in electrical conductivity caused by adding silica nanoparticles to ferroelectric liquid crystals was reported by Chaudhary et al [162] whereas an increase in conductivity by a factor of 1.4 was detected by Kaur et al [163] in similar systems.

The ion trapping effect in nematic liquid crystals doped with cobalt oxide nanoparticles was reported by Vashney et al [164]. An increase in electrical conductivity of nematic liquid crystals caused by doping them with quantum dots was observed by Shcherbinin and Konchina [134], and by Seidaliir et al [165].

Ferroelectric nanoparticles due to their spontaneous polarization are considered very promising ion trapping agents [83] and references therein. For the same reason they are also prone to aggregation. A comparative study of physical properties (including fractal analysis of aggregates and dielectric spectroscopy data as a function of temperature) of nematic and ferroelectric liquid crystals doped with paraelectric and ferroelectric BaTiO3 nanoparticles was reported by Al-Zangana et al [166]. An interesting finding presented in [166] is a non-monotonous behaviour of the electrical conductivity of liquid crystals doped with BaTiO3 nanoparticles as a function of the nanodopant concentration. In addition, the electrical conductivity of liquid crystal dispersions was greater than that of undoped liquid crystals [166]. The trapping of ions by BaTiO3 nanoparticles in bent-core liquid crystals and in antiferroelectric liquid crystals were reported by Kumar et al [167] and by Lalik et al [168], respectively.

Magnetic nanomaterials can enhance the sensitivity of liquid crystals to magnetic fields and result in intriguing magneto-optical effects [see reviews 60–62 for more detail]. They can also modify dielectric and electrical properties of liquid crystals. Improved dielectric and electro-optical properties of ferroelectric liquid crystals doped with nickel ferrite nanoparticles were reported by Sharma et al [169]. The decrease in the imaginary part of the dielectric permittivity observed in ferroelectric liquid crystal nanocomposite suggests the ion capturing behaviour of magnetic nanodopants. The non-monotonous behaviour of dissipation factor as a function of the concentration of nickel zinc ferrite nanoparticles in nematic liquid crystals was observed by Jessy et al [170]. This type of behavior was explained in terms of the competition between ion releasing and ion capturing processes in the studied systems [170]. A non-monotonous increase in the electrical conductivity of nematic liquid crystals doped with zinc ferrite nanoparticles was also detected by Pandey et al [171]. The capturing of ions by nickel zinc ferrite magnetic nanoparticles dispersed in blue phase chiral nematic liquid crystals resulted in the reduced power consumption as was reported by Jessy et al [172], γ-Fe2O3 magnetic nanoparticles were also utilized to mitigate the problem of image sticking and improve an image quality of liquid crystal displays as was shown by Ye et al [173], Gao et al [174], and Liu et al [175].

Carbon nanotubes are characterized by very high values of their mechanical strength, thermal, and electrical conductivity. In addition, when dispersed in liquid crystals, they are prone to form aggregates and networks [41–46] and references therein. Because of this, the electrical conductivity of liquid crystal materials doped with carbon nanotubes strongly depends on their concentration. According to Tomylko et al [176], different stages of aggregates and networks of carbon nanotubes in nematic liquid crystals can be identified by measuring optical and electrical properties of such composite materials. Because of the percolation effect, the electrical conductivity of liquid crystals doped with carbon nanotubes can increase by several orders of magnitude [176] (see also the paper by Ibragimov et al [177]). A substantial increase in the conductivity anisotropy and conductivity ratio observed in smectic liquid crystals doped with functionalized carbon nanotubes by Cetinkaya et al [178] is promising for applications such as conductivity switch (see also the report by Basu and Garvey [179] reporting an effective insulator-to-conductor transition in liquid crystal-carbon nanotube nanocomposites). To broaden the functionality of liquid crystal nanocomposites, carbon nanotubes can be combined with magnetic nanoparticles. Studenyak et al [180] reported a non-additive conductivity of nematic liquid crystals doped with magnetite and carbon nanotubes. The percolation driven increase in the conductivity of nematic liquid crystals doped with carbon nanotubes and magnetite was recently described by Dalir et al [181]. It should be noted that in the case of percolation phenomena, the electron conductivity of carbon nanotube-based network dominates...
over the ionic conductivity of a liquid crystal host. To observe the effects of carbon nanotubes on the ionic conductivity of liquid crystals, the concentration of nanodopants should be very small (\(\sim 10^{-4}\) wt/wt or less). DC conductivity of ferroelectric liquid crystals doped with multiwall carbon nanotubes was evaluated using cells of different thickness (5 \(\mu\)m and 12 \(\mu\)m) in the report by Shukla et al [182]. Possible applications of such soft nanocomposites in the field-sequential colour displays were also described by Shukla et al [183]. The observed nearly one order of magnitude increase in the DC conductivity of a thicker cell compared to that of a thinner cell indicates an importance of such measurements. In addition, the measured DC conductivity of both cells was a slowly increasing function of the concentration of nanotubes [182]. This finding suggests the presence of ion releasing processes in the studied composites. An increase in the ionic conductivity of liquid crystals doped with functionalized multiwall carbon nanotubes was also discussed by Patro et al [184]. Interestingly, the suppression of the screening effect due to the ion trapping phenomenon in nematic liquid crystals doped with carbon nanotubes was described by Singh et al [185].

Other types of carbon-based nanoobjects in liquid crystals have also been studied. The ion trapping effect in nanocomposites made of ferroelectric liquid crystals and nanosized diamond particles was observed by Agrahari et al [186] whereas Pandey et al [187] reported a non-monotonous dependence of the electrical conductivity as a function of the concentration of carbon quantum dots in nematic liquid crystals. A nearly two-fold decrease in the ionic conductivity of nematic liquid crystals (E7) caused by mixing them with a small fraction of graphene nanoparticles was discussed by Hadjichristov et al [188, 189] and by Marinov et al [190]. At the same time, no significant changes in the electrical conductivity of nematic liquid crystals (5CB) as a result of doping them with graphene oxide nanoflakes were found by Yadav et al [191]. A decrease in the impedance of nematic liquid crystals-graphene oxide nanocomposites was reported by Ozgan et al [192]. Basu et al designed a hybrid liquid crystal cell with a graphene layer serving as ion trapping electrode [193], and explored 2D nanomaterials made of hexagonal boron nitride and resulting in the reduction of ionic effects and improvements in electro-optics of nematic liquid crystals [194, 195].

An ongoing and very active research into the properties of liquid crystals doped with nanomaterials advances science and technology of such materials thus enabling a wide range of applications. It is interesting to discuss how existing and emerging applications of liquid crystals-nanoparticles composites can be affected by ions. It will be discussed in the next section of this paper.

Applications of liquid crystals doped with nanoparticles: the role of ions

Ions in thermotropic liquid crystals are involved in nearly any application relying on the electric-field induced reorientation of mesogenic molecules. Because nanomaterials affect the behavior of ions in liquid crystals, the consideration of such nanoparticle-induced ionic effects is very important from a practical perspective. Moreover, ions in liquid crystals are also of importance to fundamental science. In this section, we will mention some selected examples of ion-related science and applications of liquid crystals doped with nanomaterials (figure 7).

Complex electro-hydrodynamic effects in liquid crystals have been studied for decades. A recent paper by Eber et al provides a very detailed overview of such effects [196]. The presence of ions in liquid crystals is required to observe various electrically induced patterns. A very incomplete list of recently discovered intriguing effects include the formation of 3D particle-like dissipative solitons, called 'director bullets,' and soliton-induced liquid crystal enabled electrophoresis reported by Li et al [197–199]. Interesting properties of dynamic dissipative solitons in liquid crystals were also reported by Shen and Dierking [200, 201], and by Aya and Araoka [202].

As mentioned earlier, liquid crystal materials can control the self-assembly of nanomaterials (interested readers are referred to excellent reviews [28, 32, 33, 38, 39]). This process strongly depends on the surface

Figure 7. Some selected examples of science and applications of liquid crystals doped with nanomaterials.
boundary conditions defining the alignment of liquid crystalline molecules on the surface of nanoparticles. A report by Mundoor et al [203] offers new insights into the tunability of the orientation of shape-anisotropic nanoparticles in nematic liquid crystals achieved by changing the ionic content in the bulk of liquid crystals and near the surface of nanodopant. The dependence of the surface anchoring strength and anchoring type on the surface charge and bulk concentration of ions in nematic liquid crystals was recently analyzed by Everts and Ravnik [204]. It should be noted that the effects of ions on the surface anchoring were also discussed in earlier publications [113, 205–207] and references therein.

A unique versatility of liquid crystal defects offers exciting opportunities to explore fundamental concepts of topology. Interested readers are referred to comprehensive reviews by Kleman and Lavrentovich [208], Mušević [209], and Smalyukh [210] for more detail. Recent publications revealed a direct relevance of ions to topological defects in liquid crystals. Sasaki et al [211] stabilized umbilical defects by doping nematic liquid crystals with an ionic impurity. As a result, scientists achieved a large-scale self-organization of reconfigurable topological defect networks (umbilics in a square arrangement) without annihilation [211]. We can envision that ions in liquid crystals can also affect the manipulation of 2D and 3D defects in thermotropic liquid crystals by means of electric field reported recently by Ferris et al [212] and Susser et al [213]. Interestingly, topological defects can be electrically charged and can alter the surface and bulk distribution of ions in liquid crystals as was shown by Everts and Ravnik in a series of papers [214–216]. Nanomaterials can stabilize topological defects in liquid crystals (a detailed overview of this facet of liquid crystal nanoscience can be found in paper by Gudimalla et al [73]). One important application of lattices of topological defects in liquid crystals is related to the production of structured light [211, 217]. Structured light refers to the generation and application of custom light fields carrying spin angular momentum and orbital angular momentum [217]. Because of the responsiveness of liquid crystals to external stimuli, they are very suitable materials for the design of planar optical elements controlling multidimensional parameters of light [218]. Recent studies show a great potential of liquid crystal materials to advance an emerging field of flat optics [219, 220]. That’s why reconfigurable plasmonic and metamaterials made of multifunctional liquid crystals are receiving increasing attention [221–224].

Liquid crystal displays utilizing the dynamic light scattering effect caused by electrohydrodynamic instabilities in nematic liquid crystals are known since the 1960s [225–227]. Recently, this effect found very promising applications in the development of dynamic shutters and smart windows [15–19, 101, 228–230].

An understanding of sources of ion generation and ionic phenomena in liquid crystals doped with nanomaterials is critical for the development of applications discussed in this section. The knowledge of nanoparticle-induced ionic effects in liquid crystals will result in much broader range of applications of nanomaterials in liquid crystal technology. Therefore, it is important to discuss current challenges and future research directions in this vibrant research field.

Selected unsolved problems and future research directions: a perspective
This section of the review needs the following disclaimer. The choice of unsolved problems in any particular field of research is always biased by a person who makes the selection. Therefore, readers should treat the presented list of existing problems as an incomplete one and are encouraged to add new items to it.

Improved experimental techniques to measure ions in liquid crystals doped with nanomaterials
Regardless the type of nanomaterials dispersed in liquid crystals, existing experimental reports indicate that nanodopants should be considered an important source of ion generation and ion trapping in liquid crystals as was discussed in previous sections. An understanding of possible sources of ions and mechanisms of ion generation and ion trapping in liquid crystals doped with nanomaterials can be achieved by combining adequate experimental methods and suitable physical models. Karabawi et al discussed non-trivial nature of measuring a DC conductivity of liquid crystals and offered an elegant solution how to extract a true value of DC conductivity [115]. Similarly, an apparently simple evaluation of the concentration of ions in plain liquid crystals and the same liquid crystals doped with nanomaterials is not that simple [115, 117]. An analysis of existing publications reveals a broad range of the measured values of the concentration of ions even in the same liquid crystals. The fact that ions in liquid crystals can originate from different sources along with their low concentration and high sensitivity to external factors make an apparently straightforward electrical measurement a non-trivial and a tricky task. Very often the evaluated values of the concentration of ions in liquid crystals vary depending on the cell thickness [112, 182, 231, 232]. As a rule, the number of the reported experimental datapoints is typically very limited (2–3 different cell thicknesses). Therefore, the type of the dependence of the concentration of ions on the cell thickness (monotonous, non-monotonous, etc.) and physical origin of the observed behaviour are often not discussed. The presence of alignment layers makes the evaluation of the ion density in liquid crystals even more challenging problem [112, 113, 115]. The development of models focused on computing the concentration of ions is highly desirable. Even though existing models can account for the presence of alignment layers
[112, 113, 115], there are important yet overlooked factors needed to be considered. Experimental results reported by independent research groups indicate that substrates of the liquid crystal cell can either capture ions or enrich liquid crystals with ions [102–106, 193, 194, 233–235]. A reasonable quantitative explanation of the observed dependence of the concentration of ions on the cell thickness can be found in papers [106, 138, 140]. According to [106, 138, 140], the competition between ion-releasing and ion-capturing processes depends on the cell thickness, and, if not considered, can result in an incorrect evaluation of the DC electrical conductivity and concentration of ions in liquid crystals [117]. Recent conference paper [117] highlights an important practical implication: experimental measurements of the electrical conductivity and concentration of ions should be performed by using several identical cells and varying their thickness. Measurements performed at a single value of the cell thickness do not provide enough information about ionic processes in such systems.

Uncovering sources of ionic contamination of nanomaterials
Nanomaterials in liquid crystals acting as ion generating objects can be contaminated with ions prior to dispersing them in a mesogenic host. As was already mentioned in previous sections, ionic contamination of nanomaterials can result in very unusual ionic effects in liquid crystals. The identification of sources of ionic contamination of nanomaterials is a very important problem. In general, sources of ionic contamination of nanomaterials can be either intrinsic (for example, self-dissociating nanomaterials generating ions) or extrinsic (for example, uncontrolled ionic contamination as a result of synthesis, storage or handling of nanomaterials). Important information about sources of ionic contamination can be obtained by performing electrical characterization of nanomaterials dispersed in a liquid dielectric such as heptane (prior to dispersing them in liquid crystals). So far, this extra step is not popular among experimentalists. We believe it could shed light on possible sources of ionic contamination of nanomaterials and, therefore, should not be skipped. It should also be noted that external factors such as temperature, UV-light and high electric fields can become a source of ionic contamination. Some possible thermal effects are briefly discussed below.

Temperature-induced ion trapping and ion generation
Important information about ion trapping and ion generation processes can be obtained by studying temperature dependence of electrical properties of liquid crystals doped with nanomaterials. In the majority of the reported cases, the concentration of mobile ions in liquid crystals doped with nanoparticles increases as their temperature increases as a result of the temperature induced release of ions [129, 139]. A very interesting effect when the concentration of mobile ions in liquid crystals doped with nanoparticles decreases as their temperature goes up, was recently modelled [139]. This temperature-induced ion trapping effect has yet to be discovered experimentally. If found, it would enable applications of tunable liquid crystal devices operating in harsh environment. We expect that this effect can be observed in highly polar nematic liquid crystals doped with nanomaterials characterized by relatively low dielectric permittivity such as polymer nanoparticles [139].

Ion trapping and ion generation resulting in charging
Both ion trapping and ion generation processes can result in the charging of the surface of nanoparticles. It can be expected that in some cases the charged surface can result in both positive and negative charges (it must be true for ferroelectric nanomaterials). In other cases, the surface of nanoparticles can be charged either positively or negatively, or both positively and negatively (charges are distributed in a patch-like way). Even though electrophoretic motion of microparticles in liquid crystals was studied by several research groups [236–238], in the case of nanomaterials in liquid crystals these charging processes are still poorly understood. Musevic and his collaborators showed that silica nanoparticles in nematic liquid crystals 5CB are electrically charged [239, 240]. Interestingly, the total charge of nanoparticle strongly depends on its size and rapidly decreases as nanoparticle gets smaller [239, 240]. Some ferroelectric nanomaterials can also be charged [241]. The charging of ferroelectric and other types of nanoparticles in liquid crystals is an uncharted field of research. It is an established experimental fact that under certain conditions defects in liquid crystals can trap nanoparticles. It was shown that defects can be electrically charged [214–216]. The trapping of nanoparticles can be considered as interesting mechanism of the defect charging. Future studies will definitely broaden our understanding of physical and chemical mechanisms involved in charging phenomena at nanoscale.

Improved theoretical and computational models
We already discussed the simplest model describing the ion capturing and ion releasing effects in liquid crystals doped with nanomaterials (tables 1–4, figures 5, 6). Those results were obtained using linear rate equations accounting for processes of ion generation and ion trapping in liquid crystals doped with nanomaterials and placed between substrates. As was already discussed in recent papers [123, 125, 138], the utilized linear equations should be considered as a reasonable approximation. A relatively low concentration of mobile ions in liquid crystals (on the order of $10^{18}$–$10^{21}$ m$^{-3}$) is a major physical factor enabling the applicability of these equations.
In short, as a result of this small concentration of ions in liquid crystals, (i) the Bjerrum length (the distance at which electrostatic energy of two charges equals their thermal energy) is shorter than the average distance between ions; and (ii) the surface coverage of alignment layers and nanomaterials is very small thus justifying the use of this approximation. To account for much broader range of concentration of ions in liquid crystals, a more rigorous approach based on Poisson-Boltzmann equation should be developed \[242–244\]. In addition, it is very important to consider several types of dominant ions in liquid crystal nanocolloids.

Even though the discussed current challenges and future research directions were chosen in a very subjective way, there is a hope that its discussion may encourage some readers to find solutions and to expand this exciting research field. Figure 8 provides a schematic relationship between different aspects of future research directions.

**Summary**

Even though ions in molecular liquid crystals have been studied since the early 1960s, our current understanding of nanoparticle-induced ionic phenomena in liquid crystals including the origin of such effects is very incomplete. On the one hand, the ion capturing and ion releasing effects in liquid crystals doped with nanomaterials are known for more than 10 years. On the other, little is known about physical and chemical mechanisms responsible for nanoparticle-enabled ion trapping and ion generation in liquid crystals. Because of the high level of complexity of the studied systems, numerous factors are involved. They result in a great variability of the obtained results. At the same time, this variability offers new opportunities. For example, ion trapping nanomaterials can enable DC driving modes of advanced liquid crystal devices characterized by a low power consumption while ion-generating nanomaterials in liquid crystals are promising for the development of smart windows and light shutters.

An interplay between ions, nanoparticles, nanostructured surfaces, and defects in liquid crystals along with their sensitivity to external stimuli opens the door to new concepts in science and technology. Collective efforts aimed at uncovering nanoparticle-enabled ionic effects in liquid crystal materials broaden our knowledge of the complex behavior of ions in such systems. Recent progress in our understanding of ionic phenomena in liquid crystals doped with nanoparticles, reviewed in this paper, is essential for future advances in liquid crystal-aided nanoscience and nanotechnology. In the future, this understanding will be further improved by combining new experimental techniques suitable to measure ions in liquid crystals doped with new nanomaterials and predictive theoretical and computational models accounting for ionic effects in liquid crystal nanocolloids. It will lead to a much broader range of applications of nanomaterials in liquid crystal technology. Without exaggeration, the obtained knowledge of how to control ions in liquid crystals by means of nanoparticles can become a game-changer in the rapidly evolving field of liquid crystal nanoscience and advanced materials composed of liquid crystals.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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