Research Article

Nanoparticle-Enabled Ion Trapping and Ion Generation in Liquid Crystals

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Nowadays, nanomaterials in liquid crystals and their possible applications in the design of tunable, responsive, and wearable devices are among the most promising research directions. In the majority of cases, all liquid crystal based devices have one thing in common; namely, they are driven by electric fields. This type of device driving can be altered by minor amounts of ions typically present in liquid crystal materials. Therefore, it is very important to understand how nanodopants can affect ions in liquid crystals. In this paper, a recently developed model of contaminated nanoparticles is applied to existing experimental data. The presented analysis unambiguously indicates that, in general, nanomaterials in liquid crystals can behave as a source of ions or as ion traps. Physical factors determining the type of the nanoparticle behaviour and their effects on the concentration of ions in liquid crystals are discussed.

1. Introduction

Thermotropic liquid crystals are widely used in the design of tunable electro-optical devices. They include liquid crystal displays (LCD) [1], tunable lenses [2], filters [3], wave plates [3], retarders [3], diffractive optical elements [4], optical shutters [5, 6], and smart windows [7], to name a few. In the majority of cases, all of them are driven by electric fields [8]. Ions, normally present in liquid crystals in small quantities, can alter the performance of liquid crystals [9–12]. Typically, liquid crystal devices such as LCD and tunable optical elements (filters, retarders, etc.) utilize the electric field effect when the applied electric field reorients liquid crystal molecules. This type of liquid crystal based applications considers ions a nuisance because of many negative side effects caused by ions in liquid crystal devices (image sticking, image flickering, reduced voltage holding ratio, overall slow response) [9–12]. There are also electro-optical devices relying on ions in liquid crystals (optical shutters and smart windows) [5–7]. That is why an understanding of possible sources of ion generation in liquid crystals is very important [13].

Ions in molecular liquid crystals can be generated in different ways: (i) the dissociation of neutral molecules in

the bulk of liquid crystals (these dissociating species can be inherently present or added intentionally) [10–12, 14, 15]; (ii) ionic impurities as chemicals left over from the chemical synthesis [11, 16–18]; (iii) chemical degradation of liquid crystals [19]; (iv) ionic contaminants originated from the glue [20] and from the alignment layers [21–25] of the liquid crystal cell; (v)-(vii) ions generated by means of ionizing radiation [26, 27], through electrochemical reactions [28–30], and by relatively high electric fields [31–33].

Recently, 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 [[37, 38] and references therein]. From perspectives 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 reviewed in a recent paper [[39] and references therein]. Different research groups reported that various types of nanomaterials (metal [39–43], semiconductor [35, 44], dielectric [34, 36, 45], ferroelectric [46–51] magnetic [52], and carbon-based [53–56]) changed the concentration of ions in liquid crystals in different ways. Despite the variety of existing experimental results on ions

and nano-objects in liquid crystals, they can be broadly categorized into the following groups: (i) papers reporting the decrease in the concentration of ions in liquid crystals (the ion trapping regime); (ii) publications presenting the 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) [39]. A very important finding is that the same type of nanomaterials dispersed in different liquid crystals can result in different regimes (ion trapping or ion generation) [34, 39, 53]. An elementary model of these regimes was recently proposed and developed in a series of papers [57–61]. 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 [57–61]. However, the origin of the ionic contamination of nanomaterials remains practically unexplored.

In this paper, the aforementioned model of contaminated nanoparticles in liquid crystals is applied to existing experimental results with the aim of shedding some light on the nature of ionic contamination of nanodopants and ion generation/ion trapping in liquid crystals doped with such nanomaterials.

2. Elementary Model

To simplify the discussion, consider liquid crystals containing some mobile ions and nanoparticles contaminated with the same type of fully ionized ionic species. These ionic species are characterized by their volume concentration $n^+ = n^- = n$. The discussion of contaminated nanomaterials is needed to account for the possibility of experimentally observed ion trapping and ion generation regimes [57–61]. If contaminated nanoparticles are dispersed in liquid crystals, interactions between ions and nanoparticles will result in the change of the total concentration of mobile ions in liquid crystal/nanoparticle colloids. In short, a fraction of ionic contaminants can leave the surface of nanoparticles thus enriching the liquid crystal host with ions. This process can be considered as nanoparticle-enabled ion generation in liquid crystals. The reverse process, namely, the trapping of mobile ions by the surface of nanoparticles, also takes place. The competition between these two processes will result in the steady state characterized by a constant concentration of mobile ions in liquid crystals doped with nanoparticles ($dn/dt = 0$). In the simplest case, the ion releasing (or ion generation) process can be associated with the desorption of ions from the surface of nanoparticles, while the ion trapping process can be described by the adsorption of ions onto the surface of nanoparticles. As a result, the concentration of mobile ions in this system is governed by the following rate equation [62]:

$$\frac{dn}{dt} = -k_a^{NP} n_{NP} A_{NP} \sigma_S^{NP} n \left(1 - \Theta_{NP}\right)$$
$$+ k_d^{NP} n_{NP} A_{NP} \sigma_S^{NP} \Theta_{NP} \quad (1)$$

The first term of (1) describes the adsorption of ions onto the surface of nanoparticles (ion trapping process), and the second term accounts for the ion desorption from the surface of nanoparticles (ion generation process). In (1), n is the concentration of mobile ions; t denotes time; A_{NP} is the surface area of a single nanoparticle (for simplicity, consider spherical nanoparticles characterized by their radius R_{NP}; in this case $A_{NP} = 4\pi R_{NP}^2$ and this parameter determines the dependence of the concentration of mobile ions in liquid crystal nanocolloids on the size of nanoparticles); n_{NP} is the volume concentration of nanoparticles (in many practical cases the weight concentration of nanoparticles ω_{NP}, which is related to the volume concentration as $n_{NP} \approx \omega_{NP}(\rho_{LC}/\rho_{NP})(1/V_{NP})$ (V_{NP} is the volume of a single nanoparticle, and $\rho_{LC}(\rho_{NP})$ is the density of liquid crystals (nanoparticles)), is preferred); σ_S^{NP} is the surface density of all adsorption sites on the surface of a single nanoparticle; Θ_{NP} is the fractional surface coverage of nanoparticles defined as $\Theta_{NP} = \sigma_{NP}/\sigma_S^{NP}$ (σ_{NP} is the surface density of adsorption sites on the surface of nanoparticles occupied by ions); k_a^{NP} is the adsorption rate constant; and k_d^{NP} is the desorption rate constant. The aforementioned physical parameters (σ_S^{NP}, k_a^{NP}, and k_d^{NP}) are material-dependent and, for a given system under study (liquid crystals-nanoparticles), are considered as constants.

The applicability and limitations of (1) to compute the concentration of mobile ions in liquid crystals doped with nanoparticles were recently discussed in papers [63–65]. It should be stressed that (1) is an approximation which is reasonably applicable to describe ions in molecular liquid crystals. In a general case, a more rigorous approach utilizing Poisson-Boltzmann equation should be considered [66–69].

In the steady-state regime ($dn/dt = 0$), typically achieved in experiments, (1) should be solved along with the conservation law of the total number of ions expressed by

$$n_0 + n_{NP} A_{NP} \sigma_S^{NP} \nu_{NP} = n + n_{NP} A_{NP} \sigma_S^{NP} \Theta_{NP} \quad (2)$$

where n_0 is the initial concentration of ions in liquid crystals and ν_{NP} is the contamination factor of nanoparticles. The contamination factor of nanoparticles accounts for their ionic contamination and equals the fraction of the adsorption sites on the surface of nanoparticles occupied by ionic contaminants prior to dispersing nanodopants in liquid crystals [70, 71].

The ionic contamination of nanoparticles is a key factor enabling the possibility of different regimes, namely, the ion trapping regime (Figure 1, solid and dashed-dotted curves), the ion generation regime (Figure 1, dashed curve), and no change regime (Figure 1, dotted curve).

The switching between these regimes is controlled by the aforementioned contamination factor of nanoparticles ν_{NP}. The ion trapping regime is observed if $\nu_{NP} < \nu_{NP}^C$ ($n_0 > n_C$), the ion generation regime is reached if $\nu_{NP} > \nu_{NP}^C$ ($n_0 < n_C$), and no change regime is achieved if $\nu_{NP} = \nu_{NP}^C$ ($n_0 = n_C$), where ν_{NP}^C is the critical contamination factor of nanoparticles defined as $\nu_{NP}^C = n_0 K_{NP}/(1 + n_0 K_{NP})$ and n_C is the critical concentration of ions written as $n_C = \nu_{NP}/K_{NP}(1 - \nu_{NP})$
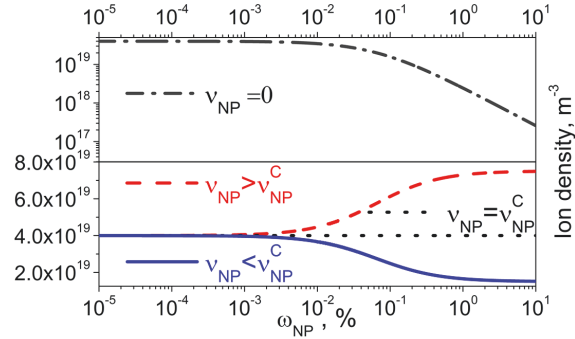
FIGURE 1: Liquid crystals doped with nanoparticles: the ion density as a function of the weight concentration of nanodopants, ω_{NP}. The case of 100% pure ($\nu_{NP} = 0$) nanoparticles in liquid crystals is represented by dashed-dotted curve (ion trapping regime). Different regimes of contaminated nanoparticles in liquid crystals are also shown: ion generation (or ion releasing) regime (dashed curve, $\nu_{NP} = 1.5 \times 10^{-3}$); ion trapping (or ion capturing) regime (solid curve, $\nu_{NP} = 3 \times 10^{-4}$); no change regime (dotted curve, $\nu_{NP} = \nu_{NP}^{C} = 8 \times 10^{-4}$). Physical parameters used in simulations: $\sigma_{S}^{NP} = 10^{18} \, m^{-2}$, $K_{NP} = k_{a}^{NP}/k_{d}^{NP} = 2 \times 10^{-23} \, m^{3}$, $n_{0} = 4 \times 10^{19} \, m^{-3}$, $R_{NP} = 10 \, nm$, and $\rho_{NP}/\rho_{LC} = 3.9$.

FIGURE 2: The concentration of ions in liquid crystals doped with TiO$_2$ nanoparticles as a function of the weight concentration of nanodopants. The presented model (solid curve) is in good agreement with reported experimental data points (squares) [34]. Physical parameters used in simulations: $\sigma_{S}^{NP} = 0.8 \times 10^{18} \, m^{-2}$, $K_{NP} = 1.6 \times 10^{-23} \, m^{3}$, $n_{0} = 1.98 \times 10^{19} \, m^{-3}$, $\nu_{NP} = 4.35 \times 10^{-4}$, $R_{NP} = 25 \, nm$, and $\rho_{NP}/\rho_{LC} = 3.9$.

and $K_{NP} = k_{a}^{NP}/k_{d}^{NP}$ (Figure 1) [57]. Both ion trapping and ion generation regimes are more pronounced if higher concentrations of nanoparticles are used (Figure 1). It should be noted that the dispersion of 100% pure nanoparticles in liquid crystals results in the ion trapping regime only (Figure 1, dashed-dotted curve). Another very important feature of the behaviour of contaminated nanoparticles in liquid crystals is an existence of the saturation effect. Upon increasing the concentration of nanoparticles in liquid crystals, the concentration of mobile ions increases or decreases towards its saturation level given by the critical concentration $n_{C} = \nu_{NP}/K_{NP}(1 - \nu_{NP})$ (Figure 1, solid and dashed curves). On the contrary, the dispersion of 100% pure nanoparticles ($\nu_{NP} = 0$) in liquid crystals does not exhibit the saturation effect (Figure 1, dashed-doted curve).

3. Case Study: TiO$_2$ Nanoparticles in Liquid Crystals [34]

The effects of titanium dioxide nanoparticles on the concentration of mobile ions in nematic liquid crystals were recently reported by Shcherbinin and Konshina [34]. By dispersing TiO$_2$ nanoparticles (Plasmotherm, Moscow) in commercially available nematic liquid crystals (ZhK1282, NIOPIK, Moscow) the ion releasing regime was observed (Figure 2, squares) [34]. These results indicate that, in the considered case, nanoparticles in liquid crystals act as a source of ions. In other words, nanoparticles, upon their dispersion in liquid crystals, increase the total concentration of mobile ions (Figure 2). According to the model described in the previous section, this increase in the concentration of ions can be caused by the ionic contamination of nanoparticles. By applying (1)-(2) of the presented model, a very good
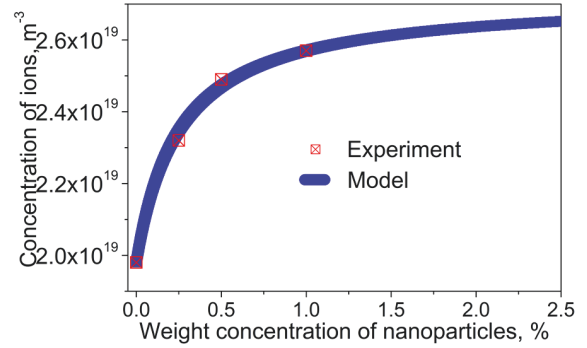
agreement between theoretical predictions and experimental results can be achieved (Figure 2). Interestingly, a minor level of the ionic contamination of nanoparticles ($\nu_{NP} = 4.35 \times 10^{-4} \ll 1$) is enough to increase the concentration of ions in liquid crystals (Figure 2). Given its small values, this uncontrolled ionic contamination of nanoparticles can easily happen during the preparation and handling of these nanomaterials.

By adding the ionic surfactant (cetyltrimethylammonium bromide, abbreviated as CTABr) to the same nematic liquid crystals (ZhK1282), the total concentration of ions was increased by nearly one order of magnitude [34]. Dispersing the same TiO$_2$ nanoparticles in these, enriched with ions, liquid crystals, the ion trapping regime was achieved [34]. To analyse these experimental results, we can consider two dominant types of ionic species in ZhK1282 liquid crystals. Pristine liquid crystals (prior to doping them with CTABr surfactants and nanoparticles) are characterized by dominant ionic species of the concentration n_{1}. Liquid crystals doped with surfactants are characterized by two dominant types of ionic species, namely, n_{1} and n_{2}. In the regime of low surface coverage of nanoparticles ($\nu_{NP} \ll 1$ and $\Theta_{NP} \ll 1$), these two dominant types of ionic species can be treated independently. As a result, the dispersion of nanoparticles in liquid crystals increases the concentration of ions n_{1} (Figure 2) and decreases the concentration of ionic species n_{2} (Figure 3) (Table 1).

The ion trapping regime, corresponding to the effects of nanoparticles on the concentration of ions n_{2}, is shown in Figure 3. As can be seen from Figure 3, there is a very good agreement between theoretical predictions based on the model of contaminated nanoparticles ((1)-(2)) and reported experimental data. As expected, TiO$_2$ nanoparticles were not contaminated with ionic species n_{2} prior to dispersing them in liquid crystals ($\nu_{NP} = 0$). As a result, the only possibility is an experimentally observed ion trapping regime (Figure 3).

TABLE 1: The concentration of ionic species in liquid crystals doped with TiO$_2$ nanoparticles [34].

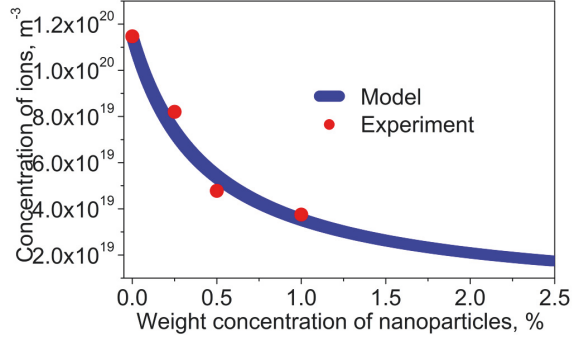| Weight concentration of TiO$_2$, %	n_1, m^{-3} (LC1)	$n_1 + n_2$, m^{-3} (LC2)	n_2, m^{-3} (LC2)
0	1.98x10^{19}	1.345x10^{20}	1.147x10^{20}
0.25	2.32x10^{19}	1.0525x10^{20}	8.2x10^{19}
0.50	2.49x10^{19}	7.275x10^{19}	4.78x10^{19}
1.00	2.57x10^{19}	6.325x10^{19}	3.755x10^{19}

FIGURE 3: The concentration of ions in liquid crystals doped with TiO$_2$ nanoparticles as a function of the weight concentration of nanodopants. The presented model (solid curve) is in good agreement with reported experimental data points (circles) [34]. Physical parameters used in simulations: $\sigma_S^{NP} = 2 \times 10^{18} \, m^{-2}$, $K_{NP} = 3.65 \times 10^{-24} \, m^3$, $n_0 = 1.147 \times 10^{20} \, m^{-3}$, $\nu_{NP} = 0$, $R_{NP} = 25 \, nm$, and $\rho_{NP}/\rho_{LC} = 3.9$.

FIGURE 4: The concentration of "slow" ions in liquid crystals doped with CdSe/ZnS quantum dots as a function of the weight concentration of nanodopants. The presented model (solid curve) is in good agreement with reported experimental data points (circles) (Table 2 of paper [35]). An inset shows the same dependence calculated at different sizes of quantum dots. Physical parameters used in simulations: $\sigma_S^{NP} = 10^{18} \, m^{-2}$, $K_{NP} = 10^{-26} \, m^3$, $n_0 = 0 \, m^{-3}$, $\nu_{NP} = 3.379 \times 10^{-3}$, $R_{NP} = 3 \, nm$, and $\rho_{NP}/\rho_{LC} = 5.091$.

4. Case Study: CdSe/ZnS Core/Shell Nanoparticles in Liquid Crystals [35]

The behaviour of ionic impurities in nematic liquid crystals (ZhK1289, NIOPIK, Moscow) doped with CdSe/ZnS core/shell quantum dots was studied by Shcherbinin and Konshina [35]. Prior to dispersing them in liquid crystals, quantum dots were functionalized with trioctylphosphine oxide (TOPO) ligands. Shcherbinin and Konshina reported that functionalized quantum dots dispersed in liquid crystals enriched the liquid crystal host with a new type of ions. These ions ("slow" ions as called in paper [35]) were not present in liquid crystals prior to mixing them with nanodopants. Thus, functionalized quantum dots were considered a primary source of these ions. It was found that the concentration of "slow" ions was a linear function of the weight concentration of nanodopants in liquid crystals [35]. This linear dependence can also be obtained by means of (1)-(2) (steady-state regime) and assuming the following conditions: $n_{NP}K_{NP}A_{NP}\sigma_S^{NP} \ll 1$ and $K_{NP}n \ll 1$. In this case, the concentration of mobile ions in liquid crystals doped with nanoparticles can be written as

$$n \approx n_0 + n_{NP}A_{NP}\sigma_S^{NP}\nu_{NP} \qquad (3)$$

As can be seen, (3) is a linear function of the concentration of nanoparticles. Again, a good match between reported experimental data and the proposed model is achieved (Figure 4).
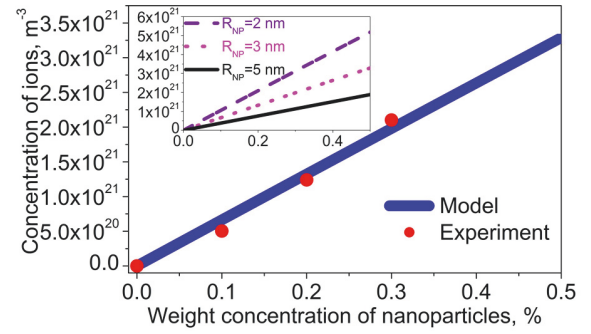
An inset (Figure 4) also shows the effect of the nanoparticle size on the concentration of ions in liquid crystals. In (1)–(3) this size effect is caused by the surface area of nanoparticles, A_{NP}. In the case of spherical nanodopants, $A_{NP} = 4\pi R_{NP}^2$. According to Figure 4 (inset), at the same concentration of nanoparticles, the ion releasing effect is greater if smaller nanoparticles are used. Additional details can also be found in recently published papers [63, 65].

5. Case Study: Cu$_7$PS$_6$ Nanoparticles in Liquid Crystals [36]

An increase in the electrical conductivity of 6CB nematic liquid crystals through doping them with Cu$_7$PS$_6$ nanoparticles was recently reported by Kovalchuk and coworkers [36]. An interesting feature of the observed effect was the saturation of the increase in the electrical conductivity at higher concentrations of nanoparticles [36].

The concentration of mobile ions n can be estimated through

$$\lambda = q\mu n \qquad (4)$$

where λ is the measured electrical conductivity, q is the charge of a single ion, and μ is its mobility [10–12]. As a result, the obtained values of the concentration of mobile ions in 6CB liquid crystals doped with Cu$_7$PS$_6$ are in a very good
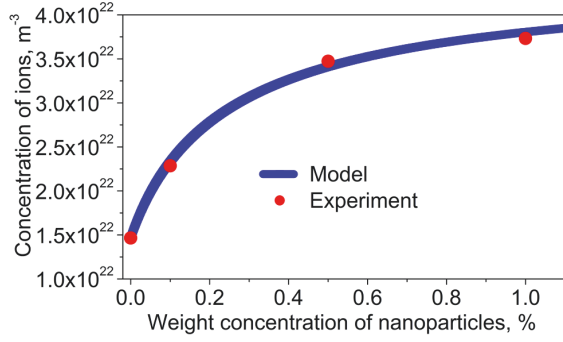
FIGURE 5: The concentration of ions in liquid crystals doped with Cu_7PS_6 nanoparticles as a function of the weight concentration of nanodopants. The presented model (solid curve) is in good agreement with reported experimental data points (circles) [36]. Physical parameters used in simulations: $\sigma_S^{NP} = 7 \times 10^{18} \, m^{-2}$, $K_{NP} = 10^{-23} \, m^3$, $n_0 = 1.466 \times 10^{22} \, m^{-3}$, $\nu_{NP} = 0.3075$, $R_{NP} = 58.5 \, nm$, $\rho_{NP}/\rho_{LC} = 4.907$, $q = 3.2 \times 10^{-19} \, C$, and $\mu = 10^{-10} \, m^2/V \times s$.

agreement with theoretical predictions based on (1)-(2) of the model of contaminated nanoparticles (Figure 5).

In the case shown in Figure 5, nanoparticles act as a source of ions in liquid crystals, thus increasing the electrical conductivity of the system under study. The concentration of ions in 6CB liquid crystals doped with Cu_7PS_6 nanoparticles as a function of the nanoparticle concentration exhibits a monotonous increase $n(\omega_{NP})$ towards the saturation level. In the framework of the proposed model, this saturation level is given by the critical concentration of ions, $n_C = \nu_{NP}/K_{NP}(1 - \nu_{NP})$. In the considered example, $\nu_{NP} = 0.35$, $K_{NP} = 10^{-23} \, m^3$, and thus $n_C = 4.44 \times 10^{22} \, m^{-3}$. Relatively high values of the contamination factor ν_{NP} can indicate that, in the case of Cu_7PS_6 nanoparticles, ionic contaminants are inherently present in these nanoparticles. In other words, the origin of these ions can be associated with the chemical structure of Cu_7PS_6 nanoparticles and the possibility of their self-dissociation rather than with ionic contaminants originated during their preparation and handling. The dissociation of Cu_7PS_6 nanoparticles resulting in the generation of copper ions can be reasonably expected for this type of materials [36].

6. Conclusions

According to existing experimental reports, nanoparticles in liquid crystals can behave either as ion traps or as ion generating objects. If nanodopants are 100% pure, the ion trapping regime is the only possible outcome of their dispersion in liquid crystals. On the contrary, liquid crystals doped with contaminated nanoparticles can exhibit three different regimes, namely, the ion trapping regime, ion generation regime, and no change regime. Both ion trapping and ion generation regimes are characterized by the saturation effect (Figure 1, solid and dashed curves, and Figures 2, 5). This effect, absent in the case of 100% pure nanoparticles in liquid crystals (Figure 1, dashed-dotted curve), is quantitatively described by the critical concentration of ions,

$n_C = \nu_{NP}/K_{NP}(1 - \nu_{NP})$. Thus, the ionic contamination of nanodopants is an important physical quantity, enabling the type of the achieved regime (ion trapping or ion generation) and leading to the saturation effect. In the framework of the model of contaminated nanomaterials, this ionic contamination is quantified by means of the dimensionless contamination factor ν_{NP} ($0 \leq \nu_{NP} \leq 1$). The predictions of this model are in very good agreement with reported experimental results (Figures 2–5). In addition, an analysis of experimental results using the model of contaminated nanoparticles can shed some light on the origin of ionic contaminants.

The ionic contamination of nanomaterials can originate from different sources during their production and handling. For example, it can be a minor fraction of ionic contaminants left over from the chemical synthesis. In the considered case study (Figure 4), trioctylphosphine oxide (TOPO) ligands are prone to uncontrolled contamination (including ionic contaminants) during the chemical synthesis [72]. As a result, once quantum dots functionalized with TOPO are dispersed in liquid crystals, ionic contaminants can be released in the bulk of the liquid crystal host. This assumption is also consistent with recent findings reported by Urbanski and Lagerwall [42]. In the case of uncontrolled ionic contamination of nanodopants, the values of the contamination factor are typically relatively low. Interestingly, even such small values of the contamination factor ($\nu_{NP} \ll 1$, on the order of 10^{-3} -10^{-4} or less, see Figures 1–4) are enough to alter the concentration of mobile ions in liquid crystals.

Ionic contaminants can also be inherently present in nanoparticles. In this case, the value of the contamination factor is relatively high ($\nu_{NP} \approx 0.1 - 1$). An example of this, inherent ionic "contamination," includes self-dissociating nanomaterials. Such nanomaterials can dissociate and generate ions in liquid crystals (Figure 5).

In general, contaminated nanoparticles should be considered as an important source of ion generation in liquid crystals. In addition, under certain conditions, they can act as ion traps. Further studies are needed to understand physical-chemical mechanisms governing these processes and identities of ionic contaminants. In the long run, this understanding will enable numerous applications of liquid crystals doped with nanomaterials.

Data Availability

All data generated or analysed during this study are included in this article (Table 1, Figures 1–5).

Conflicts of Interest

The author declares that there are no conflicts of interest.

Acknowledgments

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