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Theoretical Aspect of Nanonematic Composite: Energy Functional and Threshold Voltage

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In the present paper, we formulate a mathematical theory for dielectric anisotropy and threshold voltage of the nematic liquid crystal (NLC). We have also observed the influence of zinc oxide nanoparticles (ZONPs) on the threshold properties of the nanonematic suspension. Nanonematic suspension show changed threshold properties in comparison to the pure nematic liquid crystals. In addition to this, the effective dielectric displacement vector of composite material is calculated. We have proposed a theoretical formula for the effective dielectric anisotropy of nanonematic composite material. The equilibrium energy condition under the given constraints gives the modified threshold voltage formula and the calculated threshold voltage by this formula is found to be in accord with the experimental results.

Keywords Dielectric anisotropy; nematic liquid crystals; threshold properties; zinc oxide nanoparticles

1. Introduction

Liquid crystals (LCs) possess many unique physical, optical, and electrooptical properties. Thus, they are extremely important materials for numerous technical applications and also very interesting for many investigations in basic research [1–3]. Recently, the field has been enriched by the introduction of liquid crystalline colloids, i.e., colloidal systems in which the dispersive medium is a liquid crystal. The modifications in the physical properties of the LCs, by doping with nanoparticles, have received much attention from the viewpoint of enhanced performance of the LC [2] electrooptical devices. Several research groups have been reported on heterogeneous LC suspension using the ferromagnetic particles [3], ferroelectric nanoparticles, fullerene C-60, the nanoparticles of metals such as Pd, Ag, and Ag/Pd, and inorganic nanoparticles of MgO [2–4].

In 1969, compared with piezoelectricity in solids, R. B. Meyer introduced the concept of curvature electricity in the field of LCs, that is, the electric charge induced by the

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flexoelectric effect [4, 5]. Recently, comparing the structure of zinc oxide (ZnO) crystal and LCs, Tong et al. [6] applied the way of treating flexoelectric effect in LCs to the ZnO nanowire, and successfully explained the electricity generating mechanisms of ZnO nanowire when being bent by an exterior force [6–8]. Very recently, in 1970, Brochard and de Gennes [7] pointed out that if the colloidal particles possess a permanent magnetic moment, then the orientation of the local moments and the nematic director would be coupled.

As a piezoelectric crystal with wurtzite structure, ZnO can be considered as a slab made up of a number of planes of Zn^{2+} and O^{2-} ions stacked alternatively along its polar axis [9]. The two important characteristics of the wurtzite structure are the noncentral symmetry and polar surfaces. However, this ionic model results in a macroscopic field perpendicular to the polar surfaces, resulting in a normal dipole moment and spontaneous polarization along the C -axis as well as divergence in surface energy [8]. Both theoretical and experimental studies show that nonzero spontaneous polarization along the C -axis is still a bulk property of the ZnO crystal [8, 9]. The zinc oxide nanoparticles (ZONPs) have attracted great interest in recent years. Due to their excellent chemical and physical properties, ZONPs have a wide range of applications in piezoelectric transducers, photonic crystals, photodetectors, photodiodes, light emitting devices, transparent conductive films, etc. [10].

Keeping these facts in mind, we want to investigate the role of ZONPs in the modification of various terms in the energy functional of the ZONP-doped liquid crystalline system. This would give rise to some new interesting physical effects. The piezoelectric strain of the ZONPs may change the order parameter dielectric anisotropy, threshold voltage, and other properties of the LC. Here, we formulate a mathematical theory of dielectric anisotropy and threshold voltage of the nematic liquid crystal (NLC). The piezoelectric effect of ZONPs can be attributed for the reduction of electrostatic energy of suspension [3, 11, 12]. Our theory predicts that the NLC doped with piezoelectric nanoparticles may significantly affect the properties of the LC. This analysis also develops the understanding of the threshold behavior of the composite material used for display and other electrooptical devices [12–14].

2. Theory

To calculate the free energy functional of the system, let us consider a uniform colloidal suspension of the piezoelectric ZONPs and NLC host. The suspension confined between parallel plates was considered at $z = 0$ and $z = L$ and was subjected to planar anchoring, i.e., under the $E = 0$ condition the molecular long axes are lying in the x -axis. They were also subjected to an electric field parallel to the Z -axis. It is assumed that the deformation occurs in a plane $n = (\cos\theta, 0, \sin\theta)$, where θ is the angle between the director n and x -axis, and we assume that distortion depends only on one dimension with $\theta = \theta(z)$ [15]. The schematic diagram of the director profile is shown in Fig. 1.

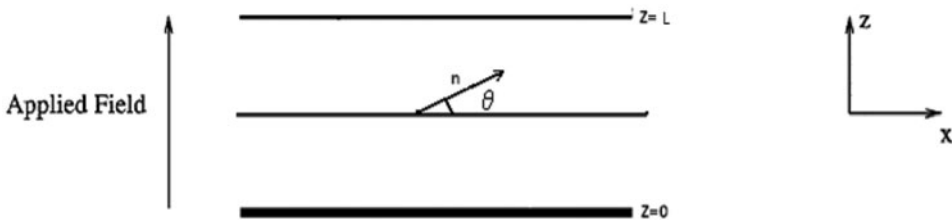


Figure 1. Schematic director profile for the nematic liquid crystal when subjected to planar anchoring and electric field perpendicular to director.

Therefore, the energy functional of the suspension is given by [16]

$$U = \int F_{\text{Total}} Dr. \quad (1)$$

This energy functional must be the sum of several contributions which are as follows:

(1) **The elastic distortion energy density:** It is the sum of splay, twist, and bend elastic energies [17, 18] and may be written as follows:

$$Fn = 1/2 K_{11} (\nabla \cdot \underline{n})^2 + 1/2 K_{22} (\underline{n} \cdot \nabla x \underline{n})^2 + 1/2 K_{33} (\underline{n} x (\nabla x \underline{n}))^2, \quad (2)$$

where the terms K_{ii} are the elastic constants and \underline{n} is the nematic director.

(2) **The dielectric energy density:** For a pure NLC system, it is given by [17, 18]

$$Fd = -1/2 \varepsilon_0 \varepsilon_{\text{II}} E^2 - 1/2 \varepsilon_0 \varepsilon_a \sin^2 \theta E_z^2, \quad (3)$$

where $\varepsilon_a = \varepsilon_{\text{II}} - \varepsilon_{\perp}$ is the dielectric anisotropy of the NLC system.

This was all about the pure NLC but when we add piezoelectric nanoparticles in the pure LC, it will affect some elastic as well as polarizing properties of the system. To study the complete effect of the nanoparticle doping on the energy functional of the pure system, one has to see the energies associated with the nanoparticles. In this respect, the first term that should get proper attention is polarization energy that can be computed as follows.

The total polarization P of the nanoparticles is the sum of the spontaneous polarization P^{SP} and the piezoelectric polarization P^{PZ} induced by the strain. In the linear regime, the strain is directly proportional to the applied electric field [19]

$$\partial \varepsilon = d \partial E, \quad (4)$$

where d = coefficient of piezoelectricity, $\partial \varepsilon$ = strain, and ∂E = electric field. The piezoelectric polarization δP^{PZ} is related to the strain as [20]

$$\delta P_i^{\text{PZ}} = \sum_j e_{ij}^{\text{PZ}} \varepsilon_j, \quad (5)$$

where e_{ij}^{PZ} are the components of the piezoelectric tensor. Correspondingly, the component along the C -axis in the ZnO nanocrystal is

$$\delta P_3^{\text{PZ}} = e_{33}^{\text{PZ}} \varepsilon_3 + e_{31}^{\text{PZ}} (\varepsilon_1 + \varepsilon_2), \quad (6)$$

where $\varepsilon_3 = (c - c_0)/c_0$ and $\varepsilon_1 = \varepsilon_2 = (a - a_0)/a_0$ are the relative changes of lattice constants c and a , describing strains along the c -axis and in the basal plane. When choosing the positive direction from anion to cation for nanoparticles [21, 22], the schematic diagram for a strained nanoparticle is shown in Fig. 2. Based on this analysis, the proposed induced piezoelectric polarization per unit volume due to slight bending in the ZONPs can be given by [21]

$$P^{\text{PZ}} = -2e_{11} R \sin \theta \hat{n}, \quad (7)$$

where e_{11} is a constant over the surface, A of the particle and R is the mean curvature of the particle shape.

Equation (7) is similar to the case of flexoelectric effect in LC film [8] but in the case of cylindrical bending of ZONPs with polar ends, it can be derived from the third component

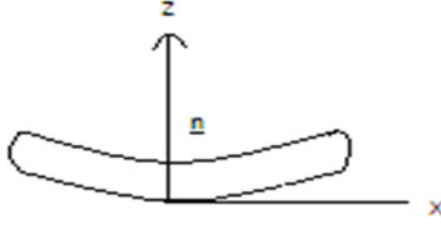


Figure 2. The strain generated in the nanoparticle after applying the electric field.

of Eq. (5) ($i = 3$) and with the help of the theory of elasticity [21, 22]. ΔP will actually have the effect of reducing the spontaneous polarization. The overall electrostatic energy will be reduced due to the slight bending of ZONPs.

Thus, the change in electrostatic energy induced by the piezoelectric polarization will be

$$\Delta F_{\text{piezo}} = \int_0^L P^{\text{PZ}} \cdot E dz. \quad (8)$$

In the absence of free electric charges, the electric field E generated by the spontaneous polarization P^{SP} will be given as

$$E = -(P^{\text{SP}}/\epsilon_0) \hat{n}. \quad (9)$$

Here, ϵ_0 is the permittivity of vacuum. By Eqs. (7) and (9), Eq. (8) becomes

$$\Delta F_{\text{piezo}} = -2e_{11} P^{\text{SP}} n \frac{\sin \theta}{\epsilon_0} \int_0^L R dz \quad (10)$$

Another form of energy associated with nanoparticles that may affect the total energy functional of composite systems is the elastic energy and this energy requires careful consideration. For the present case of ZONPs, the elastic energy of nanoparticle will only be associated with the bending deformation of nanoparticles. Thus, the elastic energy of the ZONPs can be written as

$$F_{\text{elastic}} = n \int_0^L \frac{1}{2} \eta (2R)^2 dz \quad (11)$$

where η = bending rigidity of the ZONPs. Hence, the energy functional of the particles in the suspension

$$\begin{aligned} F_{\text{nano}} = F_{\text{elastic}} + F_{\text{piezo}} = & \int_0^L C_{\text{part}} (n^{1/2} \eta \cdot (2R)^2) dz \\ & - \int_0^L (2e_{11} P^{\text{SP}} \cdot C_{\text{part}}/\epsilon_0) \cdot n \cdot \sin \theta R dz \end{aligned} \quad (12)$$

where C_{part} is the volume fraction of the nanoparticles in the suspension. Hence,

$$F_{\text{nano}} = \int [C_{\text{part}}^{1/2} \eta \cdot (2R - e_{11} P^{\text{SP}} L/\epsilon_0 \eta) 2 - C_{\text{part}} (e_{11} P^{\text{SP}} L)/2\epsilon_0 2\eta] n dz. \quad (13)$$

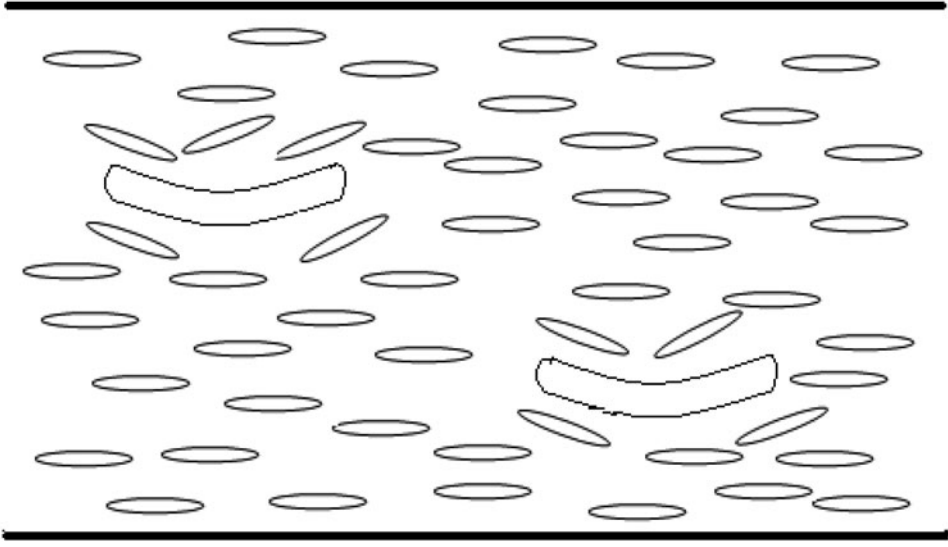


Figure 3. The strain generated due to piezoelectric effect of the nanoparticles after applying the electric field decreases the order and hence dielectric anisotropy of the suspension.

When we dope the ZONPs in the NLC, then the net energy functional of the system takes a new value to attain the equilibrium of the system. This is done by the decrease in the dielectric anisotropy of the doped system. The schematic diagram of this happening is shown in Fig. 3.

Therefore, the total energy functional of the system can be written by the sum of all energies

$$\begin{aligned}
 F = \int_0^L & 1/2 K_{11} (\nabla \cdot \underline{n})^2 + 1/2 K_{33} (\underline{n} \times (\nabla \times \underline{n}))^2 - 1/2 \epsilon_0 (\epsilon_{\perp} \\
 & + \epsilon_a^{\text{susp}} (\underline{n} \cdot \underline{E})^2) E_z^2(\theta) + \int_0^L [C_{\text{part}} \cdot 1/2 \cdot \eta \cdot n \cdot (2R - e_{11} P^{\text{SP}} L / \epsilon_0 \eta)^2 \\
 & - (C_{\text{part}} e_{11}^2 (P^{\text{SP}})^2 L^2 \cdot n) / (2\epsilon_0^2 \eta)] dz.
 \end{aligned} \quad (14)$$

If we assume that nanoparticles are small enough, then Eq. (14) becomes:

$$\begin{aligned}
 F \approx \int_0^L & \left\{ \frac{1}{2} K_{11} \cos^2 \theta + K_{33} \sin^2 \theta \right\} d^2 \theta / dz^2 \\
 & - \frac{1}{2} \epsilon_0 (\epsilon_{\perp} + \epsilon_a^{\text{susp}} \cos^2 \theta) E_z^2(\theta) dz + \text{constant}.
 \end{aligned} \quad (15)$$

In the liquid crystalline mixture, the following additive law for the effective value of dielectric anisotropy is valid, i.e.,

$$\Delta \epsilon_{\text{mixt}} = \sum_i C_i \Delta_i \epsilon. \quad (16)$$

C_i is the molar fraction of the i th mixture component taken at the reduced temperature $\tau = (T_{\text{NI}} - T) / T_{\text{NI}}$ where, T_{NI} = nematic to isotropic transition temperature [23]. With the

help of [23] and Eq. (16), now we can propose the formula for the anisotropy of nanonematic suspension as follows:

$$\varepsilon_a^{\text{susp}} = \varepsilon_a^{lc} - \lambda C_{\text{part}} f(P, \beta) \quad (17)$$

In this equation, λ is the coupling constant for dipole moment which describes the decreasing or increasing trend of dielectric anisotropy of the system. $f(P, \beta)$ represents the mathematical function of polarization and angle β . In this paper, we have not determined the value of this function but this factor should be a function of P and β . Here, P is the total dipole moment of the suspension. β is the effective angle between the long molecular axis and the dipole moment of the NLC molecule. A schematic diagram of β is given in Fig. 4 [24].

Now, the total electric displacement for the composite material

$$D = \varepsilon_o E + P_{\text{total}} = \varepsilon_o \varepsilon_{\parallel} E_z - \varepsilon_o \varepsilon_{\text{susp}}^a \sin^2 \theta E_z + P^{\text{SP}} + P^{\text{PZ}}, \quad (18)$$

where $\varepsilon_a^{\text{susp}}$ is the effective anisotropy of the system and P^{PZ} and P^{SP} have to be taken from Eq. (7) and Eq. (9), respectively. Now in the light of the boundary conditions that the suspension is under the constraints 0 to L, the threshold voltage is given by [25, 26]

$$V = \int_0^L E_z dz \quad (19)$$

Since there is cylindrical symmetry and the LC lies on the XZ plane, then $\varepsilon_{xx} = \varepsilon_{zz} = \varepsilon_{\perp} \sin^2 \theta + \varepsilon_{\parallel} \cos^2 \theta$, and $E_z = D / \varepsilon_o \varepsilon_z$ which means

$$E_z = \frac{D}{\varepsilon_o [\varepsilon_{\perp} + \varepsilon_a^{\text{susp}} \cos^2 \theta]}. \quad (20)$$

Therefore, with the help of Eqs. (17) and (19)

$$\begin{aligned} V &= \int_0^L \frac{D}{\varepsilon_o [\varepsilon_{\perp} + \{\varepsilon_a^{LC} - \lambda C_{\text{part}} f(p, \beta)\} \cos^2 \theta]} dz, \\ V &\approx \int_0^L \frac{D}{\varepsilon_o \varepsilon_{\perp}} \left[1 - \left\{ \frac{\varepsilon_a^{LC} - \lambda C_{\text{part}} f(p, \beta)}{\varepsilon_o \varepsilon_{\perp}} \right\} \cos^2 \theta \right] dz. \end{aligned} \quad (21)$$

Hence, the energy functional equation becomes

$$\begin{aligned} F &= \int_0^L \left\{ \frac{1}{2} K_{11} \cos^2 \theta + K_{33} \sin^2 \theta \right\} d^2 \theta / dz^2 \\ &\quad - \frac{1}{2} \varepsilon_o [\varepsilon_{\perp} + \{\varepsilon_a^{LC} - \lambda C_{\text{part}} f(P, \beta)\}] \cos^2 \theta E_z^2(\theta) dz. \end{aligned} \quad (22)$$

Suppose $K' \approx (K_{11} + K_{33}) \rightarrow$ for small angles

$$\begin{aligned} F &= \int_0^L \left\{ \frac{1}{2} K' d^2 \theta / dz^2 \right\} - \frac{D^2}{\varepsilon_o^2 \varepsilon_{\perp}^2} \frac{1}{2} \varepsilon_o \varepsilon_{1\perp} \left[1 + \left\{ \frac{\varepsilon_a^{LC} - \lambda C_{\text{part}} f(p, \beta)}{\varepsilon_o \varepsilon_{0\perp}} \right\} \cos^2 \theta \right] \\ &\quad \times \left[\frac{1}{1 + \left\{ \frac{\varepsilon_a^{LC} - \lambda C_{\text{part}} f(p, \beta)}{\varepsilon_o \varepsilon_{0\perp}} \right\} \cos^2 \theta} \right]^2 dz, \end{aligned} \quad (23)$$

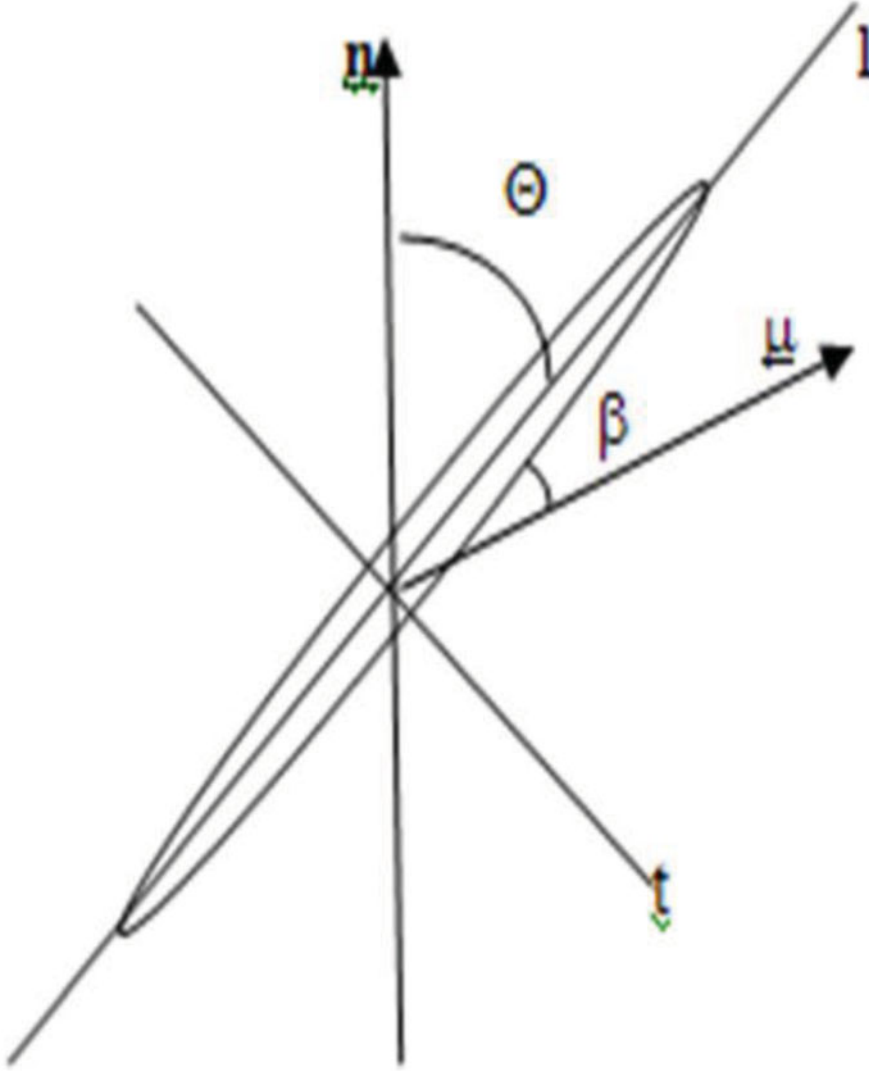


Figure 4. The schematic diagram representing angle β between the long molecular axis (\mathbf{n}) and dipole moment (μ) of nematic LC molecule. Here, \mathbf{n} denotes the direction of macroscopic nematic orientation. The subscripts l and t denotes the molecular long and short axis, respectively.

$$F = \int_0^L \left\{ \frac{1}{2} K' d^2 \theta / dz^2 \right\} - \frac{Dz}{2\epsilon_o \epsilon_{\perp}} \left[1 - \left\{ \frac{\epsilon_a^{LC} - \lambda C_{\text{part}} f(p, \beta)}{\epsilon_o \epsilon_{o\perp}} \right\} \cos^2 \theta \right] dz. \quad (24)$$

This is the expression for energy functional of the composite system. Again, from the voltage constraint of Eq. (21)

$$V = \int_0^L \frac{D}{\epsilon_o \epsilon_{\perp}} \left[1 - \left\{ \frac{\epsilon_a^{LC} - \lambda C_{\text{part}} f(p, \beta)}{\epsilon_o \epsilon_{o\perp}} \right\} \cos^2 \theta \right] dz. \quad (25)$$

We found the dielectric displacement for the suspension in simplified form, i.e.,

$$D = \frac{\varepsilon_o \varepsilon_{\perp} V}{L} \left[1 + \left\{ \frac{\varepsilon_a^{LC} - \lambda C_{\text{part}} f(p, \beta)}{\varepsilon_o \varepsilon_{o\perp}} \right\} \cos^2 \theta \right]. \quad (26)$$

The total free energy $\int_0^L F dx$ satisfies a minimization procedure which leads to the Euler–Lagrange equation in the following form [15]:

$$\frac{\partial}{\partial z} \cdot \frac{\partial F}{\partial \left(\frac{d\theta}{dz} \right)} - \frac{\partial F}{\partial \theta} = 0 \quad (27)$$

This finally gives the equation:

$$K'(\theta) \frac{d^2 \theta}{dz^2} + \varepsilon_o \varepsilon_a^{\text{susp}} \varepsilon_a E_z^2 \sin \theta \cos \theta = 0. \quad (28)$$

The solution of Eq. (28) should fulfill the boundary condition at $x = 0$ and $x = L$ and gives the approximate value for threshold voltage given as [15, 27]

$$V_{\text{th}} = \pi_{\varepsilon_{\perp}}^{\varepsilon_{\parallel}} \sqrt{\frac{K'}{\varepsilon_a^{\text{susp}}}}, \quad (29)$$

where $\varepsilon_a^{\text{sups}}$ and K' are given by Eqs. (17) and (23), respectively. Hence, the previous equation becomes

$$V_{\text{th}} = \pi_{\varepsilon_{\perp}}^{\varepsilon_{\parallel}} \sqrt{\frac{K'}{\varepsilon_a^{LC} - \lambda C_{\text{part}} f(p\beta)}} \quad (30)$$

because the strains involved in the particles due to piezoelectric effect are curvature rather than the shear and tensile strain that are involved in the usual homogeneous field crystal effects. Therefore, the guest particles disturb the molecular arrangement in the NLC matrix. Thus, the order parameter and dielectric anisotropy of the system decrease and β increases. Hence, there is an increase in the threshold voltage of the composite material system as predicted by Eqs. (29) and (30).

3. Experimental Evidence

To illustrate the effect, we have performed dielectric measurements on two samples. Two NLCs have been chosen such that one is with a highly polar LC, i.e., 5CB [28] and the other with a weakly polar LC, i.e., D6AOB [29]. We use the ZONPs [11, 28] to make the nanonematic suspension. The data were taken for different concentrations of nanoparticles with 5CB and D6AOB. The anisotropy versus concentration graph plotted at 30°C for both set of samples is presented in Fig. 5(a). The slope of these curves is negative with a multiplier. The multiplier includes the function $f(P, \beta)$ as proposed in Eq. (17). The negative slope in this equation reflects the increase in β and decrease in total polarization P .

The threshold voltages of 5CB and D6AOB samples in pure as well as in doped form are found by the capacitive method [30]. The threshold voltage versus concentration graph is shown in Fig. 5(b). It is clear that on increasing the concentration of nanoparticles in the nematic LC samples, the threshold voltage increases as predicted by Eqs. (29) and (30). This increase in the threshold voltage is higher for the 5CB as it increases more than 100%

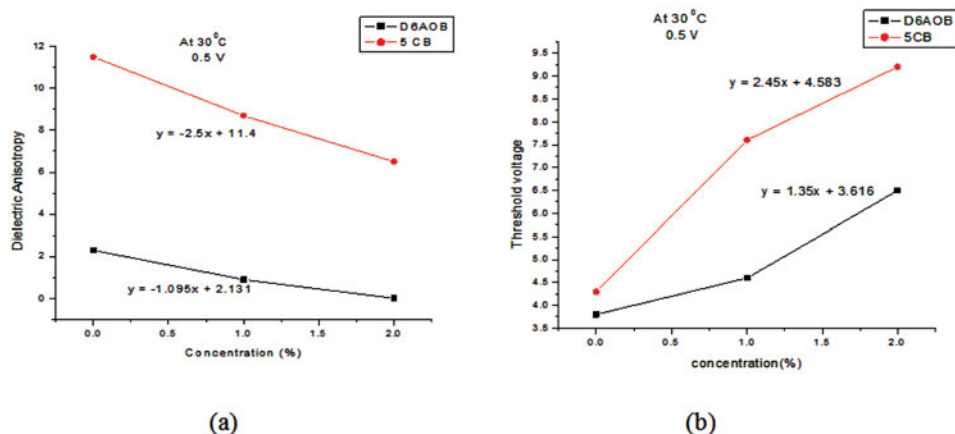


Figure 5. (a), (b). Variation of dielectric anisotropy and threshold voltage of suspension with increasing concentration of nanoparticles.

for the doped ZONPs. This behavior clearly supports the fact that the sample with the higher dielectric anisotropy shows higher effect on the threshold voltage when doped with the nanoparticle.

4. Constraints of the Proposed Theory

In our theory, we use the following limitations:

- There is no interparticle interaction.
- Each particle has permanent polarization and is subjected to piezoelectric consideration.
- The particles are only parallel to the local liquid crystal director before the applying field.
- Used nematic liquid crystal is considered to be the ideal dielectric.
- The semiconducting property of ZONPs does not have any role at room temperature in the energy functional of the suspension as the ZONPs have the wide and direct band gap of 3.37 eV.

5. Conclusion

In summary, we have examined the influence of ZONPs on the threshold properties of the nanonematic suspension. The effective dielectric displacement vector of the composite material is calculated. We proposed a theoretical formula for the effective dielectric anisotropy of nanonematic composite material. The Euler–Lagrange equation to find the equilibrium energy condition under the given constraints gives the modified threshold voltage formula. The calculated threshold voltage formula was found in accord with the experimental results. As the threshold properties are crucial for the display devices, hence deep insights toward these properties make it easy to fabricate the desired power display.

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