Large gradients of refractive index in nanosphere dispersed liquid crystal metamaterial with inhomogeneous anchoring: Monte Carlo study

Grzegorz Pawlik a,⁎, Wiktor Walasik a, Antoni C. Mitus a, Iam Choon Khoo b

a Institute of Physics, Wroclaw University of Technology, Wybrzeze Wyspianskiego 27, 50–370 Wroclaw, Poland
b Department of Electrical Engineering, Pennsylvania State University, University Park, PA 16802, USA

Abstract

We study the effect of spatial inhomogeneity of anchoring forces on real part of effective refractive index in nanosphere dispersed liquid crystal (NDLC) metamaterial at infrared frequencies using the approach of Kho et al. [1] and Monte Carlo modeling proposed recently in Ref. [2]. Local and global characterization is made using 2D maps of spatial distribution of the index, its gradients and its modulation amplitude below and above Freedericksz threshold. We find that NDLC with step-wise modulation of anchoring forces gives rise to much larger gradients and absolute values of the effective index than NDLC with modulated external electric field as well as pure nematic liquid crystal (NLC) with inhomogeneous anchoring. This indicates that the filling factor of coated spheres in NDLC is an important design parameter which tunes the effective refractive index. We find that the results are strongly dependent on wavelength in the infrared interval 2800–2900 nm. Some potential applications to molding the flow of light are briefly mentioned.

1. Introduction

Metamaterials that exhibit negative refractive indices otherwise not possible with naturally occurring materials have attracted intense interests for fundamental pursuits as well as novel applications [1,3–14]. Since the pioneering work of Veselago [3] metamaterials possessing negative refractive index have been proposed and demonstrated from microwave to optical region.

For some applications, tunability of the refractive index of the metamaterials is particularly desirable. In the case of materials containing liquid crystals the tunability is enabled by the (electrical and/or optical) field controlled birefringence of the nematic liquid crystals constituents [1,10–16]. While in the two exemplary metamaterial systems under study, namely, nanostructures with liquid crystal layers [9], and aligned nematic liquid crystals containing randomly dispersed nanospheres [1], the response of the nematic liquid crystal host to the applied external field was taken to be uniform, nonlocal inhomogeneous nematic responses are particularly important to obtain accurate quantitative description of the index modulation and other optical responses. In situations involving non-uniform applied fields, for example, in photorefractivity-mediated optical wave mixing processes [16], these detailed accurate treatments often produce new insights, and unexpected and potentially useful effects [2]. In particular, using specific spatial distributions of the electric permittivity and magnetic permeability one can curve optical space in the framework of optical transformation [17,18] and mold the flow of light in many unusual ways like, e.g., in the cases of optical black holes [19] and optical cloaking [20,21].

The emerging question is how to implement the calculated distributions of permittivity and permeability into real physical systems. It requires not only the tunability of the effective refractive index but also the control of its gradients. Until now the experimental realizations of cloaking were based either on centimeter-size Split Ring Resonators [20] or on invisibility carpet concept [21], implemented using micron-size dielectric [22] and silicon [23] nanostructures. NDLC metamaterial is well suited for this goal. Its effective refractive index at point \( \tilde{r} \) for wavelength \( \lambda \) and for some state of polarization depends, first of all, on two nonlocal and one local factors. They are (i) spatial distribution of anchoring forces \( x(\tilde{r}_W) \) at the cell’s walls \( \tilde{r}_W \), (ii) spatial distribution of external electric field \( E(\tilde{r}) \) and (iii) on local molecular orientation \( \Omega \), represented, e.g., by a set of three Euler’s angles:

\[
n_n(\tilde{r}_0) = n \left\{ x(\tilde{r}_W), \left\{ \tilde{E}(\tilde{r}), \Omega(\tilde{r}_0) \right\}, \lambda, f \right\},
\]

where \( f \) denotes the filling factor of coated nanospheres in NDLC.

Recently [2] we have studied spatial inhomogeneity of refractive index \( n_n(\tilde{r}_0) \) in NDLC metamaterial and its dependence on the wavelength, homogeneous anchoring force and modulated external electric field. To this end we have employed Monte Carlo...
simulation technique [24] which has been demonstrated to be well suited for modeling NLC properties, e.g., for chess-board NLC cell [25] and electro-optic phenomena [26]. These quantitative calculations have also enabled us to identify parameter sets that optimize the field induced index modulation and other useful metamaterial properties. In particular we have shown that spatially modulated external electric field gives rise to large local and global modulation of the amplitude of refractive index.

While these studies have elucidated in detail the role of inhomogeneous electric field $E(\vec{r})$, the anchoring forces were homogeneous: $\alpha(\vec{r}_0) = \alpha_0$. Thus the question, how inhomogeneity effects of anchoring influence the optical responses of NDLC metamaterial, remains open.

The aim of this paper is to calculate and analyze spatial inhomogeneities of refractive index resulting from spatial inhomogeneity of anchoring forces in NDLC.

### 2. Nanosphere dispersed nematic liquid crystal

Consider the case of NDLC metamaterial formed by dispersing coated spheres in NLC bulk. The effective optical responses of NDLC were calculated [1] using Mie theory [27] for scattering of a single coated sphere and Maxwell Garnet mixing rule [28,29]. Effective refractive index of NDLC metamaterial depends on parameters of coated nonmagnetic nanospheres and host NLC system.

For the case where the core is a polarizant material, its permittivity is given by:

$$\varepsilon_1 (\omega) = \varepsilon_1 (\infty) \left( 1 + \frac{\omega_p^2 - \omega^2}{\omega_p^2 - \omega^2 - i \omega \gamma_1} \right),$$

(2)

where $\varepsilon_1 (\infty)$ denotes the high-frequency dielectric constant, $\gamma_1$ is damping coefficient, $\omega_p$ and $\omega_0$ are the transverse and longitudinal phonon frequencies, respectively. Negative effective permeability of the assembly of spheres at a particular wavelength is governed by bulk cavity resonance in the core. Permittivity of the shell is described by Drude model for semiconductors:

$$\varepsilon_2 (\omega) = 1 - \frac{\omega_p^2}{\omega_p^2 + i \gamma_2 \omega},$$

(3)

where $\omega_p$ denotes plasma frequency and $\gamma_2$ – the damping coefficient. The shell is responsible for negative effective permittivity. Properly chosen parameters of the shell ensure that negative effective permittivity and permeability occur simultaneously in particular interval of wavelengths.

Permittivity of NLC host for extraordinary polarized light depends on the angle $\theta$ between the (local) director axis of NLC and wave vector of incident light [15]:

$$\varepsilon_3 (\theta) = \frac{\varepsilon_{||} \cos^2 \theta + \varepsilon_{\perp} \sin^2 \theta}{\varepsilon_{||}},$$

(4)

where $\varepsilon_{||}$ and $\varepsilon_{\perp}$ are the permittivities for light polarized parallel and perpendicular to the director axis.

The effective permittivity and permeability of NDLC metamaterial calculated in electromagnetic dipole approximation are given by [1]:

$$\varepsilon_{\text{eff}} = \varepsilon_3 \left( \frac{k_1^3 + 4 \pi i na_1}{k_2^3 - 2 \pi i na_1} \right), \quad \mu_{\text{eff}} = \frac{k_1^3 + 4 \pi i nb_1}{k_2^3 - 2 \pi i nb_1},$$

(5)

where $k_1 = \sqrt{\varepsilon_{||} k_0} = \sqrt{\varepsilon_{||} 2 \pi / \lambda}$. Scattering coefficients $a_1$ and $b_1$ can be found in Ref. [27]. Effective parameters in Eq. (5) are valid only in long-wavelength limit and for a small filling factor $f$. The effective index of refraction reads

$$n_{\text{eff}} = n_{\text{eff}}^0 + i n_{\text{eff}}^p = \pm \sqrt{\varepsilon_{\text{eff}}} \mu_{\text{eff}},$$

where root with positive imaginary part should be chosen. Khoo et al. have shown [1] that for some values of the parameters of the model there is a range of wavelengths $\lambda$ and angles $\theta$ for which $n_{\text{eff}} (\lambda, \theta) < 0$. In Fig. 1 we show the dependence of $n_{\text{eff}}^0$ on $\lambda$ and $\theta$ [2]. We have used the same values of parameters as in Ref. [1].

### 3. Monte Carlo simulation

The equilibrium configurations of NDLC were sampled using Metropolis Monte Carlo simulations [24] tailored in Refs. [25,26] to lattice model of NLC with Lebwohl–Lasher effective Hamiltonian [30] and Rapini–Papoular surface term [31]:

$$H = -\xi \sum_{\vec{r}, \vec{F}} P_2 (\cos \beta (\vec{r}, \vec{F})) - E^2 \sum_{\vec{r}} P_2 (\cos \beta (\vec{r})) + \sum_{\vec{F}} x(\vec{r}_0) \sin^2 \beta (\vec{r}_0).$$

(7)

The coupling constants $\xi$ (NLC–NLC orientational interaction), $E^2$ (square of external electric field oriented along z-axis) and $x$ (amplitude of anchoring forces) are expressed in units $k_B T$, where $k_B$ denotes Boltzmann constant and $T$ – absolute temperature. $P_2$ is the second-order Legendre polynomial. $b(\vec{r}, \vec{F})$ denotes the relative angle between two molecules located at points $\vec{r}, \vec{F}$: $\beta (\vec{r}, \vec{F})$ – the angle which a molecule located at point $\vec{r}$ makes with the electric field direction and $\gamma (\vec{r}_0)$ – the angle between long axis of molecule at the wall and a fixed rubbing direction. We have simulated the system with sizes $n_x = 100$, $n_y = 50$, $n_z = 20$ for $\xi = 25$ using periodic boundary conditions along $x$ and $y$ directions. The Freedericksz transition in this system takes place in strong anchoring regime $\alpha = 50$ at the threshold $E_\alpha \approx 0.7$. More details can be found in Ref. [2].

### 4. Inhomogeneous anchoring and inhomogeneity of effective refractive index

Large gradients of refractive index are of prime interest for design of molding of the flow of light. In this paper we study a simple but important source of a strong NLC order inhomogeneity, namely one-dimensional step-wise profile of anchoring forces, which separates regions with strong and weak anchoring:

$$\alpha(x) = \begin{cases} 1, & x \leq x_0, \\ 0, & x > x_0. \end{cases}$$

(8)

see Fig. 2, where $\alpha_0 = 50$ and $x_0 = n_x / 2 = 50$. 

![Fig. 1. Map of the real part of refractive index $n_{\text{eff}} (\lambda, \theta)$.](image-url)
For electric fields $E \leq E_F$ the NLC order is close to homogeneous for $x < x_0$ and close to homeotropic for $x > x_0$. A narrow region in the vicinity of $x_0$ is an interface with large spatial gradients of liquid crystal order. This effect is clearly seen in Fig. 3, which displays a part ($40 < x < 60$) of 2D cross-section of a 3D NDLC configuration simulated for $E = 1.1E_F$.

Given the orientations of NLC molecules we have calculated the 2D maps of distribution of $n_{\text{eff}}(x, z)$ by averaging over configurations and $y$ direction following the lines of Ref. [2]. Fig. 4 shows the maps of $n_{\text{eff}}'(x, z)$ for $\lambda = 2900$ nm below and above Freedericksz threshold: $E = 0.7E_F$ and $E = 1.1E_F$. The width of the homeotropic region increases as electric field $E$ crosses the threshold $E_F$. At the same time the width of the interface decreases and, correspondingly, the gradients $\left| \frac{\partial n_{\text{eff}}(x, z)}{\partial x} \right|$ increase their value.

In experiments an inhomogeneous local index distribution can be characterized by the averaged index $\langle n_{\text{eff}}(x) \rangle$ directly related to the phase shift of light transmitted through the cell of thickness $D$ along $z$-direction [32]:

$$\langle n_{\text{eff}}(x) \rangle = \frac{1}{D} \int_0^D n_{\text{eff}}(x, z) \, dz.$$  \hspace{1cm} (9)

In Fig. 5 we show the profiles $\langle n_{\text{eff}}(x) \rangle$ for $\lambda = 2900$ nm below, at, and above Freedericksz threshold: $E = 0.7E_F$, $E_F$ and $E = 1.1E_F$. Additionally, for comparison, we present the values of $n_{\text{eff}}'$ for purely homogeneous and homeotropic phases. As $E$ increases the width of quasi-homeotropic region increases and the plot approaches the step-function plot for $x \geq 50$; at the same time the gradient $\left| \frac{\partial \langle n_{\text{eff}} \rangle}{\partial x} \right|$ becomes larger.

Local and global characteristics of an inhomogeneous distribution of refractive index depend strongly on the wavelength of the probing light [2]. Figs. 6 and 7 show the plots of $n_{\text{eff}}'(x, z)$ and the profiles $\langle n_{\text{eff}}(x) \rangle$ for $\lambda = 2820$ nm for fields $E = 0.7E_F$ and $E = 1.1E_F$. In contrast to the case of $\lambda = 2900$ nm NDLC becomes now a metamaterial: the quasi-homeotropic order, see Fig. 3, gives rise to a metamaterial with negative index $n_{\text{eff}} < 0$, while the quasi-homogeneous part – to a metamaterial with $0 < n_{\text{eff}} < 1$. Unlike the previous case, an increase of the electric field is accompanied...
by an increase of the width of the interface and a decrease of the gradients
\( \partial n_{0}^{\text{eff}} / \partial x \) in the interface domain.

Global index distribution is characterized by the amplitude of refractive index
\( D_{0}^{\text{eff}} \) directly related to the diffraction efficiency of a thin sinusoidal phase grating:

\[
\eta = \frac{I_{\text{diff}}}{I_{0}} = J_{1} \left( 2\pi D_{0}^{\text{eff}} / \lambda \right),
\]

where \( J_{1} \) denotes the Bessel function of first kind.

For \( \lambda = 2900 \) nm the amplitude reads \( \Delta (n_{0}^{\text{eff}}) \approx 2.35 \) below the threshold \( (E = 0.7E_{F}) \) and \( \Delta (n_{0}^{\text{eff}}) \approx 2.85 \) above the threshold \( (E = 1.1E_{F}) \). The amplitude is 2–3 times larger than the amplitude for NDL system with modulated electric field reported recently in Ref. [2]. For \( \lambda = 2820 \) nm the amplitudes \( \Delta (n_{0}^{\text{eff}}) \) are approximately 1.85 for \( E = 0.7E_{F} \) and 1.6 for \( E = 1.1E_{F} \). In both cases the amplitudes of the gratings for NDL are approximately five times larger than the amplitudes for pure NLC, as depicted in Figs. 8 and 9 for \( E = 1.1E_{F} \).

This effect has interesting consequences for diffraction efficiency. In what follows we discuss the case \( E = 1.1E_{F} \) and \( \lambda = 2900 \) nm. Consider first the limit of a weak \( (\eta \ll 1) \) pure NLC grating. The diffraction efficiency in this case is \( \eta \propto \left( \Delta (n_{0}^{\text{eff}}) \right)^{2} \) and the ratio of diffraction efficiencies for NDL and NLC gratings becomes

\[
\eta_{\text{NDLC}} / \eta_{\text{NLC}} = \left( \Delta (n_{0}^{\text{eff}})_{\text{NDLC}} / \Delta (n_{0}^{\text{eff}})_{\text{NLC}} \right)^{2} \simeq 20.
\]

Thus we can obtain a remarkable increase of \( \eta \) by dispersing nanospheres into NLC. The filling factor \( f \) tunes the value of diffraction efficiency. In the case when diffraction efficiency of pure NLC is not small the relation between \( \eta \) and \( \Delta (n_{0}^{\text{eff}}) \) is more complicated and is given by Eq. (10), plotted in Fig. 10 as a function of amplitude of effective refractive index and of thickness \( D \) of the medium for \( \lambda = 2900 \) nm. In this case dispersing nanospheres in NLC allows to control the width of the cell \( D \) at constant diffraction efficiency or to control the value of the efficiency for fixed \( D \).

Figs. 8 and 9 show that NDL system has much larger gradients of effective refractive index \( \partial (n_{0}^{\text{eff}}) / \partial x \) than pure NLC. Inhomogeneous anchoring also leads to gradients which are an order of magnitude larger than those reported in Ref. [2] for the case of homogeneous anchoring but modulated electric field. Fig. 11 summarizes those conclusions.
5. Discussion and conclusions

Molding the flow of light requires an effective control of large gradients of refractive index as well as a large span of the values of the index. In the case of the propagation in metamaterials based on nematic liquid crystals this is directly related to a design of a specific molecular order. In this paper we apply Monte Carlo modeling to study the effect of spatial inhomogeneity of anchoring forces on local gradients and values of refractive index in NDLC metamaterial at infrared frequencies. We find that NDLC with step-wise modulation of anchoring forces outperforms pure NLC as well as NDLC systems with modulated external field in many situations. In particular, we note the following: The optical properties of NDLC metamaterial are strongly dependent on wavelength in the infrared interval 2800–2900 nm. The gradients of effective refractive index can be tuned by changing the homogeneous electric field close to the Freedericksz threshold. The results of this paper and of Ref. [2] indicate that the filling factor of coated nanospheres becomes an important parameter for tuning the optical properties of NDLC metamaterial. Both modulated external electric field and modulated anchoring forces are important parameters for a design of molecular order required for an implementation of geometric conditions for guiding the flow of light in NDLC. The design of particular geometries for cloaking in NDLC for chosen infrared wavelengths will be published in separate papers.

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References