Optical Materials 34 (2012) 1656–1659

Contents lists available at SciVerse ScienceDirect

Optical Materials

journal homepage: www.elsevier.com/locate/optmat

Temperature dependence of refractive index gradient in nanosphere dispersed liquid crystal (NDLC) metamaterial at infrared frequencies: Monte Carlo study

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ARTICLE INFO

Article history:
Received 17 February 2012
Accepted 5 March 2012
Available online 3 April 2012

Keywords:
NDLC metamaterial
Effective refractive index
Anchoring forces
Monte Carlo simulations

ABSTRACT

Temperature dependence of spatial inhomogeneity of real part of effective refractive index in nanosphere dispersed liquid crystal (NDLC) metamaterial at infrared frequencies is studied using the approach of Khoo et al. [1] and Monte Carlo modeling proposed recently in Refs. [2,3]. We show that the temperature is an important tuning parameter for an inhomogeneous NDLC below the temperature of nematic–isotropic liquid transition.

1. Introduction

The field of metamaterials exhibiting negative values of real part of effective refractive index attracted a lot of interest during the last decade [4,5]. Various methods of fabrication of those non naturally occurring materials have been proposed. Metamaterials with large gradients of refractive index are highly desirable for purposes such as optical cloaking [6,7].

Recently we have shown [3] that the nanosphere dispersed liquid crystal (NDLC) metamaterial allows to create large gradients of the index due to a proper choice of anchoring forces at fixed temperature. The objective of this paper is the calculation and analysis of the influence of temperature on gradient of refractive index, resulting from spatial inhomogeneity of anchoring forces in NDLC.

2. Nanosphere dispersed nematic liquid crystal

The NDLC metamaterial is built of coated nonmagnetic spheres randomly dispersed in nematic liquid crystal (NLC). A sphere consists of a core and a shell. The core is made of a polaritonic material with permittivity given by:

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

where $\varepsilon_1(\infty)$ denotes the high-frequency dielectric constant, γ_1 denotes damping coefficient, ω_T and ω_L represent the transverse and longitudinal phonon frequencies, respectively. According to the effective medium theory recapitulated briefly below, the core resonance is responsible for the negative values of effective permeability of the assembly of spheres at a particular wavelength. The semiconductor shell has the permittivity described by Drude model:

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

where ω_p denotes plasma frequency and γ_2 – the damping coefficient. The shell parameters determine the values of effective permittivity. Properly chosen parameters of the shell ensure that negative effective permittivity and permeability occur in the same interval of wavelengths.

Permittivity of NLC host for extraordinary polarized light depends on the angle θ between the (local) director axis of NLC and wave vector of incident light [8]:

$$\varepsilon_3(\theta) = \frac{\varepsilon_\parallel \varepsilon_\perp}{\varepsilon_\parallel \cos^2\theta + \varepsilon_\perp \sin^2\theta}, \tag{3}$$

where $\varepsilon_\parallel$ and $\varepsilon_\perp$ are the permittivities for light polarized parallel and perpendicular to the director axis, respectively.

The optical response of a single coated sphere was calculated [1] using Mie scattering theory [9]. The effective optical parameters of NDLC metamaterial, calculated using Maxwell Garnet mixing rule [10,11], depend on parameters of coated nanospheres

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http://dx.doi.org/10.1016/j.optmat.2012.03.009

and host NLC system. In the electromagnetic dipole approximation the expressions for the effective permittivity and permeability of NDLC metamaterial read [1]:

$$\varepsilon_{eff} = \varepsilon_3\left(\frac{k_3^3 + 4\pi i N a_1}{k_3^3 - 2\pi i N a_1}\right), \quad \mu_{eff} = \frac{k_3^3 + 4\pi i N b_1}{k_3^3 - 2\pi i N b_1}, \tag{4}$$

with $k_3 = \sqrt{\varepsilon_3}k_0 = \sqrt{\varepsilon_3}2\pi/\lambda$, where λ denotes the vacuum wavelength. Scattering coefficients a_1 and b_1 can be found in Ref. [9]. Effective parameters in Eq. (4) are valid only in long-wavelength limit and for a small filling factor f. The expression for the effective refractive index reads:

$$n_{eff} = n'_{eff} + in''_{eff} = \pm\sqrt{\varepsilon_{eff}\mu_{eff}}, \tag{5}$$

where the root with positive imaginary part should be chosen. Khoo et al. have shown [1] that $n'_{eff}(\lambda,\theta) < 0$ for some values of the parameters of the model.

3. Monte Carlo simulation

The equilibrium configurations of NDLC were sampled using Metropolis Monte Carlo simulations [12] tailored in Refs. [13,14] to a lattice model of NLC with Lebwohl–Lasher effective Hamiltonian [15] and Rapini–Papoular surface term [16]:

$$H = -\xi\sum_{<\vec{r},\vec{r}'>} P_2(\cos\beta(\vec{r},\vec{r}')) - E^2\sum_{\vec{r}} P_2(\cos\beta(\vec{r})) + \sum_{\vec{r}_W}\alpha(\vec{r}_W) \times \sin^2\gamma(\vec{r}_W). \tag{6}$$

Parameter ξ (we have used $\xi = 25$ in the simulations) denotes the strength of an orientational interaction between NLC molecules, E stands for the amplitude of an external electric field oriented along z-axis (Fig. 1) and α denotes the amplitude of anchoring force. P_2 is the second-order Legendre polynomial. $\beta(\vec{r},\vec{r}')$ denotes the relative angle between two molecules located at points $\vec{r},\vec{r}'$; $\beta(\vec{r})$ – the angle between a molecule located at point $\vec{r}$ and the electric field direction and $\gamma(\vec{r}_W)$ – the angle between long axis of molecule at the wall and a fixed rubbing direction. We have simulated the system on the lattice with sizes $n_x = 100$, $n_y = 50$, $n_z = 20$ in corresponding directions, see Fig. 1, using periodic boundary conditions along x and y directions. Each lattice site is occupied by a single NLC molecule. The relation to real-space unit of length follows from the observation that in experimental setups the width of the cell in z direction constitutes a few microns. The Freedericksz transition in this system occurs, in strong anchoring regime $\alpha = 50$, at the electric field threshold $E_F \approx 0.7$. More details can be found in Ref. [2].

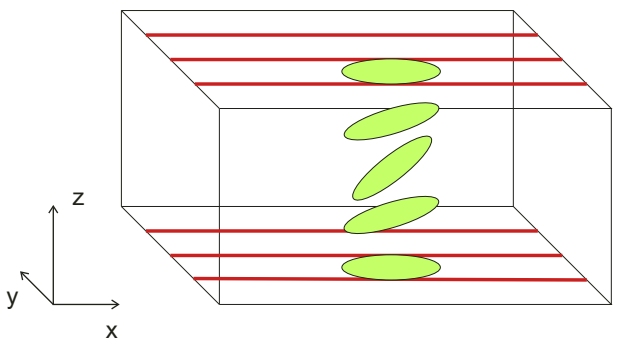

Fig. 1. Geometry of the simulated system. Electric field is along the z axis. Horizontal lines denote the direction of rubbing.

4. Temperature dependence of refractive index distribution for inhomogeneous anchoring

Large gradients of refractive index are of primary importance for the design of molding of the flow of light. Current study is restricted to a simple but important case of a one-dimensional step-wise profile of anchoring forces given by Ref. [3]:

$$\alpha(x) = \begin{cases} \alpha_0 & \text{for } x \leqslant x_0 \\ 0, & \text{for } x > x_0 \end{cases}, \tag{7}$$

where $x_0 = n_x/2 = 50$. In the region $x \leqslant x_0$ the anchoring is strong ($\alpha_0 = 50$), whereas for $x > x_0$ the anchoring is weak.

The global order in the system is characterized by order parameter S, which takes values between 0 for a completely disordered phase and 1 for a completely ordered phase. Order parameter can be inferred from the ensemble average of the second rank tensor [8]:

$$Q_{\alpha\beta} = \frac{1}{N}\sum_{i=1}^{N}\left(\frac{3}{2}\hat{n}_{i,\alpha}\hat{n}_{i,\beta} - \frac{1}{2}\delta_{\alpha\beta}\right), \quad \alpha,\beta = x,y,z, \tag{8}$$

where $\hat{n}_{i,\alpha}$ denotes the αth component of the versor describing the orientation of the long axis of the ith NLC molecule, N denotes number of molecules, and $\delta_{\alpha\beta}$ – the Kronecker delta function. The diagonalization of ensemble averaged tensor $Q_{\alpha\beta}$ yields three eigenvalues λ_+, λ_0 and λ_- ($\lambda_+ > \lambda_0 \geqslant \lambda_-$) which sum up to zero. For an uniaxial system the the following relation holds:

$$\lambda_- = \lambda_0 = -\frac{1}{2}\lambda_+. \tag{9}$$

In this paper we use $S = \lambda_+$ as the order parameter.

Monte Carlo simulations of a homogeneous system with periodic boundary conditions in all three dimensions yield the value $k_BT_c/\xi = 1.15$ for the reduced temperature of phase transition between ordered NLC and isotropic liquid, see Fig. 2. Here, k_B denotes Boltzmann constant and T – absolute temperature. The presence of an electric field and anchoring forces may have a strong impact on the character of the transition – this topic will be studied in subsequent papers. In Fig. 3 we show the 2D maps of a real part of effective local refractive index for a few values of the reduced temperature ($k_BT/\xi = 0.04$ (a), $k_BT/\xi = 0.4$ (b), $k_BT/\xi = 0.6$ (c), $k_BT/\xi = 0.8$ (d), $k_BT/\xi = 1.12$ (e)). These temperatures are related to the temperature T_c in the following way: $T = 0.035\,T_c$ (a), $T = 0.35T_c$ (b), $T = 0.52T_c$ (c), $T = 0.7T_c$ (d), and $T = 0.97T_c$ (e). We have used the wavelength $\lambda = 2875$ nm and external electric field $E = 0.7 \simeq E_F$. For sufficiently low temperatures

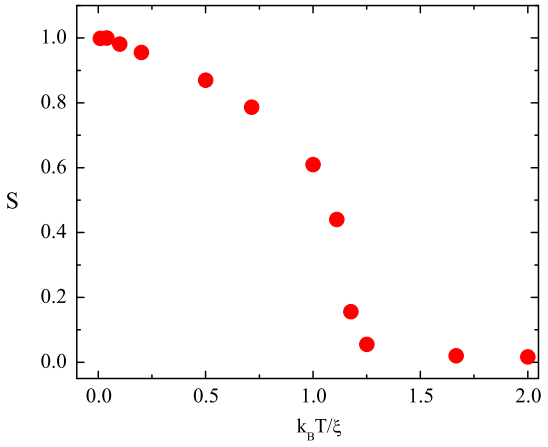

Fig. 2. Dependence of order parameter $S = \lambda_+$ on reduced temperature k_BT/ξ in a homogeneous NLC system.