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Optical Model to Describe Coherent Transmittance and Absorptance of Polymer Dispersed Liquid Crystal Film Doped with Carbon Nanotubes at Normal Interface Anchoring

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Abstract

An optical model has been developed for analyzing the coherent transmittance and absorptance of a polymer dispersed liquid crystal films doped with carbon nanotubes (CNTs) at uniform normal droplet-polymer interface anchoring. It is based on the Foldy–Twersky and anomalous diffraction approximations, Maxwell-Garnett equations, and the order parameters concept. The model allows one to analyze the electro-optical response of films depending on the film thickness, the refractive indices of the liquid crystal (LC) and the polymer matrix, the size and concentration of the LC droplets, the concentration of nanotubes, the conductivities and permittivities of the CNTs, LC and the polymer. Experimental verification of the model is performed.

1 Introduction

The polymer-dispersed liquid crystal (PDLC) films [1] consist of a polymer matrix containing liquid crystal (LC) droplets, in which the orientation of the LC molecules can be changed under the electric or magnetic fields. This allows one to control the optical response of the films. They are used in displays, optoelectronic, microelectronic, and telecommunication systems, laser devices, etc. Electrically or magnetically controlled optical response of PDLC films is based on light scattering. It does not require the use of additional polaroids in comparison with the ordinary (bulk) LC layers.

In recent years, there is an increasing interest in studying the dielectric and optical properties of composite materials based on bulk LC and PDLC films doped with carbon nanotubes (CNTs) [2-5]. This is due to additional opportunities provided by nanotubes (NTs) to form and control electro-optical response.

Currently, studies of the electro-optical response of composite PDLC-CNTs films are mainly experimental [4,5]. As far as we know, there are no theoretical optical models that allow one to describe and predict electro-optical response of the PDLC-CNTs films as function of the component parameters (LC, polymer, NTs).

In this paper, we suggest an electro-optical model for analyzing the coefficients of coherent (directional, regular) transmission (coherent transmittance) and absorption (absorptance) of a PDLC-CNTs film with a homogeneous normal interface anchoring. To determine the coherent transmittance of the film, the Foldy–Twersky approximation is used [6,7]. The optical characteristics of a single droplet of nematic LC are determined in the framework of the anomalous diffraction approximation [1,7] using the effective refractive indices of the droplet [6]. Based on the Maxwell–Garnett equations [8], a method has been developed to determine the refractive index of the polymer matrix, the effective refractive index of the LC droplets, and the threshold field of the reorientation of the director structure of LC droplets upon doping the PDLC film with NTs.

A technique has been developed for determining the volume filling factor of the film with LC droplets, volume filling factors of the LC droplets and polymer matrix doped with NTs, depending on the mass fractions of the components in the PDLC-CNTs composite. The technique is applicable to singlewall (SWCNTs) and multiwall (MWCNTs) carbon nanotubes. Experimental verification of the developed model was carried out.

2 Theory

Let us consider a PDLC film containing a polydisperse ensemble of spheroidal LC droplets with rotation symmetry relatively to small axis directed along the normal to the film. The anisometry parameter ε_a, defined as the ratio of the major axis of the droplet in the film plane to the minor axis along the normal to the film, is the same for all droplets. In the framework of the Foldy-Twersky approximation, it is possible to write the following expressions for the coherent transmittance T_c and albedo Λ of a film:

$$T_c = \exp(-\gamma_{ext} l), \quad (1)$$

$$\Lambda = Q_{scat} / Q_{ext} = Q_{scat} / (Q_{scat} + Q_{abs}), \quad (2)$$

$$\gamma_{ext} = \frac{3c_d}{4a_{ef}} Q_{ext}, \quad (3)$$

where γ_{ext} is the extinction coefficient of the PDLC film; l is the film thickness; Q_{scat}, Q_{ext} and Q_{abs} are the scattering, extinction and absorption efficiency factors for single droplet; c_d is the volume filling factor of the film with LC droplets (ratio of the volume of all droplets to the volume of film where they are distributed); a_{ef} is the effective [7] length of the minor droplet semiaxis a along the normal to the film.

Using the anomalous diffraction and effective medium approximations and based on the results of [6,7], we obtained:

$$Q_{ext} = 4\,\mathrm{Re}\,K_h(v_{ext}), \quad (4)$$

$$v_{ext} = i2ka_{ef}(m-1), \quad (5)$$

$$Q_{abs} = 2K_h(v_{abs}), \quad (6)$$

$$v_{abs} = 2\,\mathrm{Re}\,v_{ext}, \quad (7)$$

$$k = 2\pi/(\lambda m_{p+CNTs}), \quad (8)$$

$$m = m_{d+CNTs}/m_{p+CNTs}, \quad (9)$$

where K_h is the Hulst function [1],

$$K(v) = \frac{1}{2} - \frac{\exp(-v)}{v} + \frac{\exp(-v)-1}{v^2}, \quad (10)$$

m is the complex refractive index of LC droplet relative to the refractive index of polymer matrix; λ is the wavelength of the incident light in vacuum; m_{d+CNTs} and m_{p+CNTs} are the effective refractive indices of the LC droplet and polymer matrix doped with nanotubes. They are determined as follows:

$$m_{d+CNTs} = \sqrt{\varepsilon_{d+CNTs}}, \quad (11)$$

$$\varepsilon_{d+CNTs} = \varepsilon_{m+CNTs} - \frac{1}{3}\Delta\varepsilon_{LC+CNTs} SS_{d+CNTs}(E), \quad (12)$$

$$\varepsilon_{m+CNTs} = \frac{2\varepsilon_{LC+CNT,\perp} + \varepsilon_{LC+CNT,||}}{3}, \quad (13)$$

$$\Delta\varepsilon_{LC+CNTs} = \varepsilon_{LC+CNT,||} - \varepsilon_{LC+CNT,\perp}, \quad (14)$$

$$m_{p+CNTs} = \sqrt{\varepsilon_{p+CNTs}}. \quad (15)$$

In Eqs. (11)-(15), ε_{d+CNTs} and ε_{p+CNTs} are the complex effective permittivities droplet and polymer with CNTs; ε_{m+CNTs} and $\Delta\varepsilon_{LC+CNTs}$ are the average value permittivity and dielectric anisotropy of liquid crystal; $\varepsilon_{LC+CNTs,||}$ and $\varepsilon_{LC+CNTs,\perp}$ are permittivities parallel and orthogonal to director of LC; S is the molecular order parameter of LC [1]; $S_{d+CNTs}(E)$ is the order parameter of LC droplets with nanotubes, depending on the control electric field E.

To find of $\varepsilon_{LC+CNTs,||}$, $\varepsilon_{LC+CNTs,\perp}$ and ε_{p+CNTs} we used the Maxwell-Garnett approximation:

$$\varepsilon_{LC+CNTs,||} = \varepsilon_{LC,||}\left(1 + c^d_{CNTs}\left(R_{LC+CNTs,||} - 1\right)\right), \quad (16)$$

$$R_{LC+CNTs,||} = \varepsilon_{CNTs,||}/\varepsilon_{LC,||}, \quad (17)$$

$$\varepsilon_{LC+CNTs,\perp} = \varepsilon_{LC,\perp}\frac{1 + 0.5(1 + c^d_{CNTs})\left(R_{LC+CNTs,\perp} - 1\right)}{1 + 0.5(1 - c^d_{CNTs})\left(R_{LC+CNTs,\perp} - 1\right)}, \quad (18)$$

$$R_{LC+CNTs,\perp} = \varepsilon_{CNTs,\perp}/\varepsilon_{LC,\perp}, \quad (19)$$

$$\varepsilon_{p+CNTs} = \varepsilon_p\left\{1 + \frac{c^p_{CNTs}}{3}\,\frac{R_{p+CNTs,||} - 1 + 4\dfrac{R_{p+CNTs,\perp} - 1}{R_{p+CNTs,\perp} + 1}}{1 - (2/3)c^p_{CNTs}\dfrac{R_{p+CNss,\perp} - 1}{R_{p+CNTs,\perp} + 1}}\right\}, \quad (20)$$

$$R_{p+CNTs,||} = \varepsilon_{CNTs,||}/\varepsilon_p, \; R_{p+CNTs,\perp} = \varepsilon_{CNTs,\perp}/\varepsilon_p, \quad (21)$$

where $\varepsilon_{LC,||}$ and $\varepsilon_{LC,\perp}$ are the permittivities of LC; ε_p is the permittivity of polymer; $\varepsilon_{CNTs,||}$ and $\varepsilon_{CNTs,\perp}$ are the permittivities of nanotubes; c^d_{CNTs} and c^p_{CNTs} are the volume filling factors of the LC droplets and the polymer matrix with CNTs (characterizing the part of droplets and polymer matrix volume occupied by CNTs).

For droplets with normal interface anchoring, when a control field is applied along the normal to the film, using the results of [8], it is possible to write the following expression for the droplet order parameter in Eq.(12):

$$S_{d+CNTs}(E) = 1 - \exp(-e). \quad (22)$$

Here $e=E/E_c$ is the dimensionless normalized value of the control field: E_c is the critical value of the control field [9-11].

3 Results

To compare the results obtained in the framework of the developed model with the measurement data, we used the experimental dependences of the normalized transmittance $T_c^{norm}=T_c/T_c^{max}$ (T_c^{max} is maximum transmittance) on the control field E.

The measurements were carried out for PDLC films based on E7 LC and PMMA polymer without NTs and upon doping with NTs. The samples were illuminated along the normal to the film surface by a He-Ne laser at the wavelength λ=0.6328 μm. In the absence of NTs, ordinary $n_\perp$ and extraordinary $n_{||}$ refractive indices of the LC are equal to: $n_\perp$=1.52, and $n_{||}$=1.745, the refractive index of the polymer n_p=1.503. The filling factor of the PDLC film c_d=0.435. Films thickness l=50 μm. The average radius of droplets in the plane of the samples is $\langle a\rangle$=1.07 μm; effective droplet size a_{ef}=2.34 μm. The experimental and theoretical dependences of $T_c^{norm}(E)$ are presented in Figure 1. The following parameters were used in the calculation: the volume filling factor of the film with nanotubes c_{CNT}=0.03 (c^d_{CNT}=0.011, c^p_{CNT}=0.019); the electrical conductivities of LC $\sigma_{||}$=5.7×10^{-8} S/m and $\sigma_\perp$= 2.6×10^{-8} S/m. The calculations were carried out for different values of the electrical conductivities σ_p of the polymer matrix with and without nanotubes. One can see a good agreement of the theoretical and experimental results at variation of the model parameters in both cases.

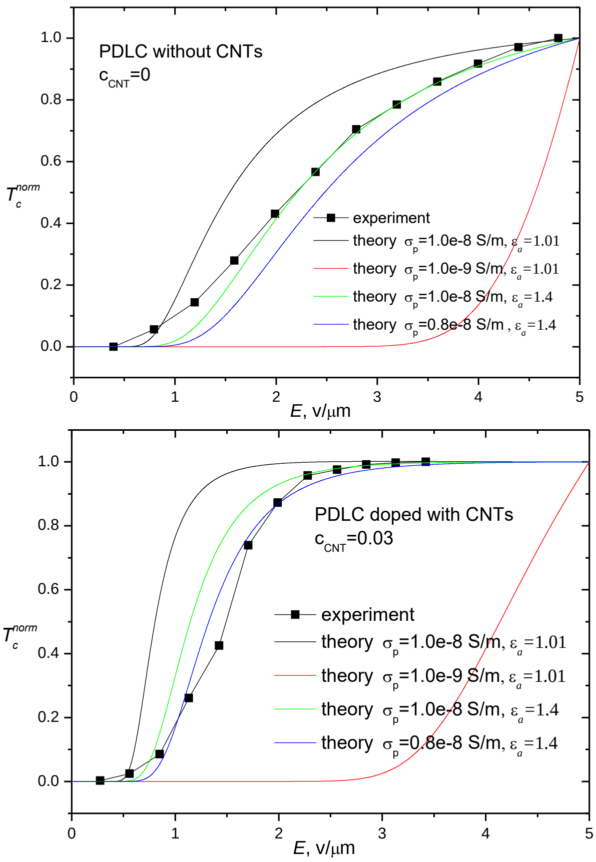

Figure 1 *Theoretical and experimental dependences of the normalized transmittance $T_c^{norm}(E)$ for the PDLC film (top) and PDLC-CNTs film (bottom) at different values of the electrical conductivities σ_p of the polymer matrix*

4 Conclusions

In the present work a model for describing the electro-optical response of PDLC-CNT films is developed. The model is validated by comparison with experiment.

Pay attention that the theory is developed for the films with a uniform normal interface anchoring. Such films are characterized by a polarization-independent coherent transmittance at normal illumination, when the control electric field is directed normally to the film. They are promising for applications in optoelectronic devices where modulation of light is required without changing its polarization state and to modulate the unpolarized light. The model can be extended to PDLC-CNT films with polarization-dependent tangential interface anchoring and the PDLC films doped with other nanoparticles: gold, ferroelectric, silica, etc.

5 References

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