AFRL-AFOSR-UK-TR-2013-0039

Effective dielectric, magnetic and optical properties of isotropic and anisotropic suspensions of ferroic nano-particles

Viktor Yuriyovych Reshetnyak

Kyiv National Taras Shevchenko University
Physics Faculty
Prospekt Glushkova, 2, Building 1
Kyiv, 03680
UKRAINE

EOARD STCU 07-8001/Project P-338a

Report Date: June 2013

Final Report for 18 October 2007 to 30 June 2013

Distribution Statement A: Approved for public release distribution is unlimited.

Air Force Research Laboratory
Air Force Office of Scientific Research
European Office of Aerospace Research and Development
Unit 4515 Box 14, APO AE 09421

REPORT DOCUMENTATION PAGE	Form Approved OMB No. 0704-0188

1. REPORT DATE ***(DD-MM-YYYY)***	**2. REPORT TYPE**	**3. DATES COVERED** *(From – To)*
24 June 2013	Final Report	18 October 2007 – 30 June 2013

4. TITLE AND SUBTITLE

Effective dielectric, magnetic and optical properties of isotropic and anisotropic suspensions of ferroic nano-particles

5a. CONTRACT NUMBER
STCU 07-8001/Project P-338a

5b. GRANT NUMBER
STCU 07-8001

5c. PROGRAM ELEMENT NUMBER
61102F

6. AUTHOR(S)

Viktor Yuriyovych Reshetnyak

5d. PROJECT NUMBER

5d. TASK NUMBER

5e. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

Kyiv National Taras Shevchenko University
Physics Faculty
Prospekt Glushkova, 2, Building 1
Kyiv, 03680 UKRAINE

8. PERFORMING ORGANIZATION REPORT NUMBER
N/A

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)

EOARD
Unit 4515
APO AE 09421-4515

10. SPONSOR/MONITOR'S ACRONYM(S)
AFRL/AFOSR/IOE (EOARD)

11. SPONSOR/MONITOR'S REPORT NUMBER(S)
AFRL-AFOSR-UK-TR-2013-0039

12. DISTRIBUTION/AVAILABILITY STATEMENT

Distribution A: Approved for public release; distribution is unlimited.

13. SUPPLEMENTARY NOTES

14. ABSTRACT The present project focuses on the theoretical study of suspensions of nano-particles of different nature (ferroelectric, ferromagnetic, multiferroic) with size of 10-100nm in isotropic and anisotropic host. We shall investigate the effect of the dispersed nano-particles on the dielectric, magnetic and optical (both linear and non-linear) properties of the suspensions depending on the nano-particles nature, size, structure and shape. To describe the electric response of suspension of ferroelectric nanoparticles in insulating non-polar (isotropic) fluid we use a theory which is based on the Fokker-Planck (Smoluchowski) equation for a rotator with electric dipole under the external electric field. We also use the dynamic equation for reorientation of ferroelectric nano-particles in the isotropic suspension under the external electric field. We developed the generalized Maxwell-Garnett type theory to study dielectric properties of a dilute suspension of ferroelectric particles in a nematic liquid crystal host. It is supposed that submicron particles do not disturb the LC alignment and the suspension macroscopically appears similar to a pure LC. We propose theoretical model for effective dielectric function of ferroelectric LC suspension. It is found that particles permanent polarisation may significantly increase the effective value of suspension dielectric function in comparison with pure LC. For more elongated particles depolarisation factor is smaller and respectively the contribution of induced polarisation of particles to the effective dielectric function is higher. We show that there are two sets of the effective permittivity tensors. One of these can be used to derive the dielectric properties like capacitances. A second set must be used in free-energy calculations to predict director profile in the cells where, for example, director field is inhomogeneous. Neglecting positional correlations of the nanoparticles we get expressions for both sets of the effective permittivity tensors. Considering Freedericksz transition in a ferroelectric LC suspension we have shown that under condition when inhomogeneous distribution of impurity ferroelectric nanoparticles is stabilized the Freedericksz transition threshold can change its value at change of applied voltage sign. We found dependence of Freedericksz transition thresholds and their asymmetry on spontaneous polarization of nanoparticles, weight concentration of nanoparticles, pattern of their spatial distribution and director anchoring energy on the cell substrates. We have shown that there is an area of parameters values, close to the experimental data, which describes experimental results for Freedericksz transition threshold in the TL205 LC cell doped with BaTiO3 ferroelectric nanoparticles. We considered different cases of collective behavior of ferroelectric nanoparticles in the LC cell and studied their influence on electric field penetrating into the cell from photorefractive substrates. For these cases we calculated spontaneous polarization which nanoparticles create and took it into account to determine electric field in the cell. We show that spontaneous polarization of nanoparticles renormalizes the principal components of dielectric function and diffraction grating wave number depending on the type of nanoparticle collective behavior. Dielectric function of the ferroelectric nanoparticles suspension in nematic LC cell with photorefractive substrates is investigated. Influence of electric field penetrating into the cell from photorefractive substrates on dielectric function of suspension is studied. We found that dielectric function of suspension depends essentially on director anchoring on the nanoparticle surface. For the cases of planar and homeotropic director anchoring we obtained analytical expressions which allow to calculate dielectric function at different values of ferroelectric nanoparticle suspension parameters. We studied the impact of ferroelectric nano-particles on two-beam coupling gain in the hybrid nematic LC cells. We established that model of strong correlation of nano-particles orientation with the LC director field is applicable for describing experimental results for gain coefficient in LC TL205 doped by BaTiO3 nano-particles. In this model gain coefficient changes its sign and becomes negative with increase of grating spacing and nanoparticle concentration. Gain coefficient depends also on mutual orientation of nano-particles long axis and LC director as well as a sign of high frequency dielectric anisotropy of ferroelectric suspension.

15. SUBJECT TERMS
EOARD, ferroelectric, ferromagnetic and multiferroic, new photorefractive effects in liquid crystal cell, new materials and systems for information processing, inorganic substrates and liquid crystal with flexo

16. SECURITY CLASSIFICATION OF:			**17. LIMITATION OF ABSTRACT**	**18, NMBER OF PAGES**	**19a. NAME OF RESPONSIBLE PERSON**
a. REPORT	**b. ABSTRACT**	**c. THIS PAGE**			John Gonglewski
UNCLAS	UNCLAS	UNCLAS	SAR	16	**19b. TELEPHONE NUMBER** *(Include area code)* +44 (0)1895 616007

Effective dielectric, magnetic and optical properties of isotropic and anisotropic suspensions of ferroic nano-particles

Project manager: Viktor Yuriyovych Reshetnyak, Professor
Phone: 380-44-5264477, Fax: 380-44-2353113, E-mail: reshet@iop.kiev.ua
Institutions: Kyiv National Taras Shevchenko University
Financing parties: USA
Operative commencement date: 01.04. 2011
Project duration: 2 years 3 months
Project technical area: Physics
Reported: 01.04.2011 – 30.06.2013
Date of submission: 24.06.13

Executive summary

Mutual influence of nano-particles and isotropic/anisotropic liquid gives rise to new and often unique physical properties. Adding of nano-sized objects in isotropic/anisotropic materials may result in significant enhancement of their electro/magneto-optic and optoelectronic properties. It is possible to increase the composite materials sensitivity to the external electric or magnetic field and even generate a new set of properties: dielectric, magnetic, mechanical, luminescence etc absent in a pure material. The idea of doping the liquid crystals with elongated ferromagnetic particles to increase the magnetic susceptibility of a liquid crystal host was first proposed by Brochard and de Gennes [1]. They have also developed the continuum theory of such systems assuming infinitely strong orientational coupling between LC molecule director and the surface of the colloidal particle. Liang and Chen [2, 3] investigated the magneto-optical behavior of ferronematics at low magnetic field, and have shown that there is no threshold voltage of the Frederiks transition. Burylov and Raikher resolved a problem of orientation of elongated particle in a uniformly distorted LC host [4], and generalized the continuum theory [1] to the finite orientational coupling between the nematic molecules and the particle surface [5]. However, they have made no attempts to find new possible effects considering different geometries of the ferronematic system. The group of Prof. Sluckin (University of Southampton, UK) in collaboration with the group of Prof. Reshetnyak (University of Kyiv, Ukraine) has extended their theoretical studies, carried out computer simulations of ferromagnetic LC suspensions in different geometries, and also predicted the inverse Frederiks effect in these systems [6-10]. The LC materials filled with ferroelectric nano-particles were developed and studied at AFRL, Dayton, Ohio (Cook et al), Kent State University (West et al), Institute of Physics (Reznikov et al) and University of Kyiv (Reshetnyak et al) in the Ukraine [11-15]. The group of Prof. Kobayashi (Liquid Crystal Institute of Tokyo University of Science, Japan) carries out experimental studies of dielectric and optical properties of nanoolloids, the group of Prof. Chen (Chaio Tung University, Taiwan) investigates the properties of ferroelectric LCs doped with ferroelectric particles.

The present project focuses on the theoretical study of suspensions of nano-particles of different nature (ferroelectric, ferromagnetic, multiferroic) with size of 10-100nm in isotropic and anisotropic host. We shall investigate the effect of the dispersed nano-particles on the dielectric, magnetic and optical (both linear and non-linear) properties of the suspensions depending on the nano-particles nature, size, structure and shape.

To describe the electric response of suspension of ferroelectric nanoparticles in insulating non-polar (isotropic) fluid we use a theory which is based on the Fokker-Planck (Smoluchowski) equation for a rotator with electric dipole under the external electric field. We also use the dynamic equation for reorientation of ferroelectric nano-particles in the isotropic suspension under the external electric field.

We developed the generalized Maxwell-Garnett type theory to study dielectric properties of a dilute suspension of ferroelectric particles in a nematic liquid crystal host. It is supposed that submicron particles do not disturb the LC alignment and the suspension macroscopically appears similar to a pure LC. We propose theoretical model for effective dielectric function of ferroelectric LC suspension. It is found that particles permanent polarisation may significantly increase the effective value of suspension dielectric function in comparison with pure LC. For more elongated particles depolarisation factor is smaller and respectively the contribution of induced polarisation of particles to the effective dielectric function is higher. We show that there are two sets of the effective permittivity tensors. One of these can be used to derive the dielectric properties like capacitances. A second set must be used in free-energy calculations to predict director profile in the cells where, for example, director field is inhomogeneous. Neglecting positional correlations of the nanoparticles we get expressions for both sets of the effective permittivity tensors.

Considering Freedericksz transition in a ferroelectric LC suspension we have shown that under condition when inhomogeneous distribution of impurity ferroelectric nanoparticles is stabilized the Freedericksz transition threshold can change its value at change of applied voltage sign. We found dependence of Freedericksz transition thresholds and their asymmetry on spontaneous polarization of nanoparticles, weight concentration of nanoparticles, pattern of their spatial distribution and director anchoring energy on the cell substrates. We have shown that there is an area of parameters values, close to the experimental data, which describes experimental results for Freedericksz transition threshold in the TL205 LC cell doped with $BaTiO_3$ ferroelectric nanoparticles.

We considered different cases of collective behavior of ferroelectric nanoparticles in the LC cell and studied their influence on electric field penetrating into the cell from photorefractive substrates. For these cases we calculated spontaneous polarization which nanoparticles create and took it into account to determine electric field in the cell. We show that spontaneous polarization of nanoparticles renormalizes the principal components of dielectric function and diffraction grating wave number depending on the type of nanoparticle collective behavior.

Dielectric function of the ferroelectric nanoparticles suspension in nematic LC cell with photorefractive substrates is investigated. Influence of electric field penetrating into the cell from photorefractive substrates on dielectric function of suspension is studied. We found that dielectric function of suspension depends essentially on director anchoring on the nanoparticle surface. For the cases of planar and homeotropic director anchoring we obtained analytical expressions which allow to calculate dielectric function at different values of ferroelectric nanoparticle suspension parameters.

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Cooperation with foreign collaborators

Travels of prof. V. Reshetnyak

to Yerevan, Armenia for participation in 14th International Topical Meeting on Optics of Liquid Crystals 2011, September 25 - October 1, 2011. At the conference V.Yu. Reshetnyak delivered two reports on scientific results of the project;

to Mainz, Germany, for participation in conference ILCC'12 (24-th International Liquid Crystal Conference), August 19 - August 26, 2012. Two talks on results of the project were given at the conference;

to Hong Kong, for participation in the 4th Workshop on Liquid Crystals for Photonics, and to Japan, Fuji Calm, for participation in the 1st Asian Conference on Liquid Crystals, December 7 - December 20, 2012. Two talks on results of the project were given at these conferences.

Travels of prof. V. Reshetnyak and prof. I. Pinkevych

for participation in 9th International Photorefractive Working Meeting (Marco Island, Florida, June 20-24, 2011) and 10th International Photorefractive Working Meeting (Long,oat Key, Florida, June 10-15, 2012). Two talks on results of the project were given at each Meeting;

for participation in 2nd Workshop on Complex Liquids at Structured Surfaces, 10-12 October 2012, Lisbon, Portugalia. One talk on results of the project was given.

for participation in 4th COINAPO Topical Meeting Ljubljana, Slovenia, December 8-9, 2011) and COINAPO Workshop (Ercolano, Italy, March 20-21 2013. Two talks on results of the project were given at these conferences.

Travel of prof. I. Pinkevych to Cambridge, United Kingdom (May 16 – June 3, 2013) to participate in the programme "The Mathematics of Liquid Crystals" in the Isaac Newton Institute for Mathematical Sciences. One talk on results of the project was given.

Publications

1. Sergii M. Shelestiuk, Victor Yu. Reshetnyak, and Timothy J. Sluckin. Phys. Rev. E **83**, 041705 (2011).
2. S. A. Basun, G. Cook, V. Yu. Reshetnyak, A. V. Glushchenko, and D. R. Evans. Phys. Rev. B **84**, 024105 (2011).
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5. V. Yu. Reshetnyak, G. Cook, I. Pinkevych, T.J. Sluckin and D. R. Evans. Asymmetric Frederiks transition in liquid crystal doped with ferroelectric nano-particles. 14th International Topical Meeting on Optics of Liquid Crystals 2011, September 25 - October 1, Yerevan, Armenia, I-32.
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7. Sergii M. Shelestiuk, Victor Yu. Reshetnyak. Effective Dielectric Function of Ferroelectric Nanosuspensions. 4th COINAPO Topical Meeting, 8-9 December, Ljubljana, Slovenia, O21.
8. I. P. Pinkevych, V.Yu. Reshetnyak, G. Cook, D. R. Evans. Two-beam energy exchange in a hybrid organic-inorganic cell containing ferroelectric nano-particles. 4th COINAPO Topical Meeting, 8-9 December, Ljubljana, Slovenia, O22.

9. Gary Cook, Dean Evans, Anatoly Glushchenko and Victor Yu Reshetnyak. Nanoparticle doped hybrid photorefractives - United States Patent 8018648, Application Date: 2008-06-12, Publication Date 2011-09-13.
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11. I. P. Pinkevych, V.Yu. Reshetnyak, G. Cook and D.R. Evans. Photorefractivity and two-beam energy exchange in a hybrid organic-inorganic cell containing ferroelectric nano-particles. 2nd Workshop on Complex Liquids at Structured Surfaces, 10-12 October 2012, Lisbon, Portugalia, Book of Abstracts.
12. I. P. Pinkevych, G. Cook, D.R. Evans, V.Yu. Reshetnyak. Two-beam energy exchange in a hybrid organic-inorganic cell containing ferroelectric nano-particles. 24th International Liquid Crystal Conference, August 19th - 24th 2012, Mainz, Germany, Book of Abstracts.
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14. Igor P. Pinkevych, Victor Yu. Reshetnyak, Gary Cook, Dean R. Evans, Dynamic holographic grating and two beam energy exchange in hybrid cholesteric liquid crystal cell. Cost Action MP0902 Composites of inorganic nanotubes and polymers, March 20-22, 2013, Ercolano, Italy, Book of Abstracts, p.22.

Prospects of future development.

Results of the project allow suggesting new isotropic and LC-based systems with nano-sized particles for information processing, dynamic recording and storage. They may also be used for the future core telecommunications technologies. After completion of the work plan the final results of the project will be presented at the international conferences and submitted for publication in the internationally recognized journals.

Leading firms, laboratories, and university centers whose scientific activities (commercial, fundamental, or both) depend upon advancements in our field:

1. Air Force Research Laboratory, Wright-Patterson Air Force Base, Dayton, OH, USA
2. Azimuth Corporation, Dayton, Ohio 45432, USA
3. Bell Laboratories, Lucent Technologies, New Jersey, USA.
4. Liquid Crystal Institute, Kent State University, Kent, Ohio, USA.
5. University of Colorado at Boulder, USA.
6. University of Colorado at Colorado Springs, USA
7. Case Western Reserve University, Cleveland, USA
8. The Pensylvania State University, Pensylvania, USA
9. University of Artois, Lens, France
10. University of Paris-Sud 11, Orsay, France
11. University of Montpellier II, France
12. Institute of Applied Physics, Darmstadt University of Technology, Darmstadt, Germany.
13. Technical University of Berlin, Berlin, Germany.
14. Universita di Ancona, Ancona, Italy.
15. Università di Bologna, Italy
16. University of Naples Federico II, Napoli, Italy
17. Dipartimento di Fizica Universita della Calabria, Rende, Italy.
18. Tokyo University of Science, Japan.
19. Centro de Investigación en Química Aplicada, Mexico
20. University of Southampton, Southampton, United Kingdom.
21. University of Manchester, Manchester, United Kingdom.
22. University of Warwick, United Kingdom
23. Yerevan State University, Yerevan, Armenia.
24. Military University of Technology, Warsaw, Poland
25. University of Ljubljana, Ljubljana, Slovenia
26. Institute of Physics, Kiev, Ukraine.
27. Kiev National Taras Shevchenko University, Kiev, Ukraine.

Project Manager For the Coordinating Institution

Approved

Reshetnyak V.Yu.	Makarets M.V. Dean of Physics Faculty Taras Shevchenko National University of Kyiv
	For the STCU
Senior Specialist	Approved
Taberko E.	<signature>
Deputy Executive Director	For the STCU
	Approved
<enter name>	<signature>

Project main idea

Mutual influence of nano-particles and isotropic/anisotropic liquid gives rise to new and often unique physical properties. Adding of nano-sized objects in isotropic/anisotropic materials may result in significant enhancement of their electro/magneto-optic and optoelectronic properties. It is possible to increase the composite materials sensitivity to the external electric or magnetic field and even generate a new set of properties: dielectric, magnetic, mechanical, luminescence etc absent in a pure material. Diluted colloids of solid inorganic ferro-, para- and superparamagnetic, ferroelectric nano-particles in LCs modify properties of liquid crystals improving device technology, in particular, enabling devices to be controlled using easily accessible electric and/or magnetic fields. These systems are important in telecommunications, flat-screen displays, optical sensors and visualizers of electric/magnetic field, switching devices, photonic band-gap meta-materials, and liquid crystal lenses. At the same time, to explain the observed effects and forecast new ones, further development of a theory is needed.

The idea of doping the liquid crystals with elongated ferromagnetic particles to increase the magnetic susceptibility of a liquid crystal host was first proposed by Brochard and de Gennes [1]. They have also developed the continuum theory of such systems assuming infinitely strong orientational coupling between LC molecule director and the surface of the colloidal particle. Liang and Chen [2, 3] investigated the magneto-optical behavior of ferronematics at low magnetic field, and have shown that there is no threshold voltage of the Frederiks transition. Burylov and Raikher resolved a problem of orientation of elongated particle in a uniformly distorted LC host [4], and generalized the continuum theory [1] to the finite orientational coupling between the nematic molecules and the particle surface [5]. However, they have made no attempts to find new possible effects considering different geometries of the ferronematic system. Recently the group of Prof. Sluckin (University of Southampton, UK) in collaboration with the group of Prof. Reshetnyak (University of Kyiv, Ukraine) has extended their theoretical studies, carried out computer simulations of ferromagnetic LC suspensions in different geometries, and also predicted the inverse Frederiks effect in these systems [6-10]. Chernyshuk, Lev and Yokoyama considered theoretically collective effects in doped nematic liquid crystals and shown these effects to be strongly dependent on the anchoring strength, particle shape and concentration [11]. The LC materials filled with ferroelectric nano-particles were developed and studied at AFRL, Dayton, Ohio (Cook et al), Kent State University (West et al), Institute of Physics (Reznikov et al) and University of Kyiv (Reshetnyak et al) in the Ukraine [12-16]. Several groups all over the world have started studying the photorefractive effect in hybrid organic- inorganic LC and LC doped with ferroelectric NPs. In particular, the group of Dr. Evans (AFRL, Dayton, Ohio, USA) and the group of Prof. Kaczmarek (University of Southampton, UK) are studying experimentally the photorefractive and non-linear optical properties of ferroelectric LC nanocolloids, the group of Prof. Kobayashi (Liquid Crystal Institute of Tokyo University of Science, Japan) carries out experimental studies of dielectric and optical properties of nanoolloids, the group of Prof. Chen (Chaio Tung University, Taiwan) investigates the properties of ferroelectric LCs doped with ferroelectric particles, the group of Prof. Čopič (J. Stefan Institute, Slovenia) studies the properties of ferroelectric LC nanocolloids by dynamic light scattering technique [14, 17-21].

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21. V.Yu. Reshetnyak, I. P. Pinkevych, G. Cook, D. R. Evans and T. J. Sluckin *Phys. Rev. E* **81**, 031705 (2010)

The present project focuses on the theoretical study of suspensions of nano-particles of different nature (ferroelectric, ferromagnetic, multiferroic) with size of 10-100nm in isotropic and anisotropic host. We plan to investigate the effect of the dispersed nano-particles on the dielectric, magnetic and optical (both linear and non-linear) properties of the suspensions depending on the nano-particles nature, size, structure and shape.

The following results are expected to be obtained in the project: a) to study the effect of size, shape, concentration and structural organization of feroic nano-particles on dielectric/ magnetic/optical properties of isotropic and anisotropic suspensions;b) to develop a generalized Maxwell-Garnett type theory for the effective dielectric/magnetic function of low concentrated ferroic LC nano-suspension will by developed; c) to clarify the role of the permanent and induced dipoles in the dielectric response of the LC suspension, and in particular its effect on asymmetrical Frederiks transition; d) to study impact of particles chaining, director and particles concentration spatial inhomogeneity on dielectric/ magnetic response of suspensions; e) to clarify conditions for individual and collective behavior of nano-particles of different nature in the suspension; f) to investigate influence of the anchoring at the particles surface on the effective dielectric/magnetic function of the suspension; g) to study impact of ferroic nanoparticles on diffraction efficiency and two-beam coupling in the hybrid LC.

In the project we plan to understand the basic physics of dielectric, magnetic and optical properties of isotropic and anisotropic suspensions of ferroic nano-particles thus advancing the knowledge about the effects that may take place in these materials. It will allow suggesting new isotropic and LC-based systems with nano-sized particles for information processing, dynamic recording and storage. Results of the project may also be used for the future core telecommunications technologies.

Technical approach

To describe the electric response of suspension of ferroelectric nanoparticles in insulating non-polar (isotropic) fluid we use a theory which is based on the Fokker-Planck (Smoluchowski) equation for a rotator with electric dipole under the external electric field E .

Let θ, φ are the polar and azimuth angles of the nanoparticle electric dipole orientation, and $f(\theta,\varphi,t)$ is the orientational distribution function for the nanoparticle dipole. Ehe Fokker-Planck equation for distribution function $f(\theta,\varphi,t)$ can be written in the form

$$\frac{\partial f(\theta,t)}{\partial t} = D\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial f(\theta,t)}{\partial\theta}\right) + E\frac{d}{\gamma}\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin^2\theta\, f(\theta,t)\right) \quad (1)$$

Using the derivative $\frac{\partial f(\theta,t)}{\partial t}$ from equation (1) s we arrive to the following equation for suspension polarization

$$\frac{\partial <P>}{\partial t} = -\frac{<P>}{\tau} + \frac{\eta d^2}{\gamma}E<\sin^2\theta> \quad (2)$$

where $\tau = (2D)^{-1}$ is the rotational relaxation time, $<\sin^2\theta>=\int_0^{\pi}\sin^2\theta f(\theta,t)\cos\theta d\theta <\sin^2\theta>$.

Supposing fluctuations of the polar angle to be small we replace $<\sin^2\theta>$ in equation (2) by approximate expression, $<\sin^2\theta>=1-<\cos^2\theta>\approx 1-\left(<\cos\theta>\right)^2 = 1-\left(\frac{<P>}{\eta d}\right)^2$, and obtain

$$\frac{\partial <P>}{\partial t} = -\frac{<P>}{\tau} + \frac{\eta d^2}{\gamma} E[1-\left(\frac{1}{\eta d}\right)^2 <P>^2] \tag{3}$$

Equation (3) can be used to calculate the displacement current

$$i_d = S\cdot\frac{\partial <P>}{\partial t}\ , \tag{4}$$

where S is a cell substrate area. In general case, system of equations (3), (4) can be solved numerically.

We consider a suspension of ferroelectric particles, with anisotropic polarizability, embedded in a LC medium, and occupying a fraction f of the total volume. We suppose that there are strong forces aligning the principal axis of a particle with the local LC director. If we define $\mathbf{P}_p$ as the mean permanent dipole moment per unit volume of particle and that these forces are sufficiently strong that we may suppose that the vectors $\mathbf{P}_p$ and the director $\mathbf{n}$ are aligned. We distinguish the local electric fields inside the particles and those inside the nematic LC. We average electric and displacement fields over volumes which are small, in the spirit of continuum theory. The key electric fields in the theory are $\mathbf{E}_{LC}$ and $\mathbf{E}_P$, the averaged electric fields in the LC and ferroelectric particles, respectively, as well as the average field $\mathbf{E}$ in the whole medium. Within a mean field theory in which the colloid particles are dilute, well-separated, and uncorrelated in position, the quantities $\mathbf{E}_{LC}$, $\mathbf{E}_P$, and $\mathbf{E}$ are related as follows:

$$\mathbf{E} = (1-f)\mathbf{E}_{LC} + f\,\mathbf{E}_P \tag{5}$$

$$\mathbf{D} = \mathbf{D}_0 + f\,\mathbf{P}_P = (1-f)\mathbf{D}_{LC} + f\,\mathbf{D}_P + f\,\mathbf{P}_P \tag{6}$$

The quantities $\mathbf{D}=\hat{\hat{\varepsilon}}\mathbf{E}$, $\mathbf{D}_0$, $\mathbf{D}_{LC}=\varepsilon_0\hat{\varepsilon}^{LC}\mathbf{E}_{LC}$, and $\mathbf{D}_P=\varepsilon_0\hat{\varepsilon}^P\mathbf{E}_P$ are defined, respectively, as the mean displacement field, the contribution to the mean displacement field resulting from induced polarization effects, the mean displacement field in the LC, and the mean-induced displacement field in the ferroelectric particles. In addition, we introduce a relevant electric field $\mathbf{E}_P^0$, which orients the permanent polarization of the particles. This differs from the electric field $\mathbf{E}_P$ in that it does not include the field due to the permanent polarization inside the particles themselves. In order to calculate the permanent polarization $\mathbf{P}_P$, we recall the strong anchoring condition that the ferroelectric particles be aligned only parallel or antiparallel to the local LC director. For suitably defined fields

$$\mathbf{E}_P^0 = \hat{T}^0\mathbf{E}_{LC}\ ,\quad \mathbf{E}_P = \hat{T}\,\mathbf{E}_{LC}\ ,\quad \mathbf{E}_{LC} = \hat{T}^{LC}\,\mathbf{E} \tag{7}$$

The quantities $\hat{T}^0$, $\hat{T}$, $\hat{T}^{LC}$ are tensors. We generalize effective medium theory of Maxwell-Garnett so that both permanent polarization of the colloidal particles and anisotropy effects are included. We thus calculate the transformation matrices by considering a host anisotropic LC medium with dielectric permittivity tensor $\hat{\varepsilon}^{LC}$, and a single spheroidal particle with dielectric permittivity tensor $\hat{\varepsilon}^P$ placed inside this medium. A key result links the mean LC electric field $\mathbf{E}_{LC}$, the mean particle electric field $\mathbf{E}_P$, and the mean polarization $\mathbf{P}_P$. This is

$$\hat{\varepsilon}^{LC}\mathbf{E}_{LC} = \left[\hat{\varepsilon}^{LC} + \hat{\hat{\mu}}\left(\hat{\varepsilon}^P - \hat{\varepsilon}^{LC}\right)\right]\mathbf{E}_P + \hat{\hat{\mu}}\,\mathbf{P}_P/\varepsilon_0 \tag{8}$$

where $\hat{\hat{\mu}}$ is the so-called depolarization tensor of a dielectric spheroid in an anisotropic medium.

Also we can write that

$$\hat{\varepsilon}^{LC}\mathbf{E}_{LC} = \left[\hat{\varepsilon}^{LC} + \hat{\mu}\left(\hat{\varepsilon}^{P} - \hat{\varepsilon}^{LC}\right)\right]\mathbf{E}_{P}^{0} \tag{9}$$

Using above mentioned equations we can find the relevant expressions for the components of the dielectric permittivity tensor $\hat{\tilde{\varepsilon}}$

$$\begin{aligned} \tilde{\varepsilon}_{\perp} &= (1-f)\,\varepsilon_{\perp}^{LC}T_{\perp}^{LC} + f\,\varepsilon_{\perp}^{P}T_{\perp}^{P} \\ \tilde{\varepsilon}_{\parallel} &= (1-f)\,\varepsilon_{\parallel}^{LC}T_{\parallel}^{LC} + f\,\varepsilon_{\parallel}^{P}T_{\parallel}^{P} + f\,\nu T_{\parallel}^{P0} \end{aligned} \tag{10}$$

and components of T- matrices

$$\begin{aligned} \hat{T}^{0} &= \left[I + \left(\hat{\varepsilon}^{LC}\right)^{-1}\hat{\mu}\left(\hat{\varepsilon}^{P} - \hat{\varepsilon}^{LC}\right)\right]^{-1} \\ \hat{T} &= \left[I + \left(\hat{\varepsilon}^{LC}\right)^{-1}\hat{\mu}\left(\hat{\varepsilon}^{P} - \hat{\varepsilon}^{LC}\right)\right]^{-1}\left[I - \nu\left(\hat{\varepsilon}^{LC}\right)^{-1}\hat{\mu}\hat{n}\hat{T}^{0}\right] \\ \hat{T}^{LC} &= \left[(1-f)I + f\,\hat{T}\right]^{-1} \\ T_{\parallel}^{0} & \frac{\varepsilon_{\parallel}^{LC}}{\varepsilon_{\parallel}^{LC} + \tilde{\mu}_{\parallel}\left(\varepsilon_{\parallel}^{P} - \varepsilon_{\parallel}^{LC}\right)} \end{aligned} \tag{11}$$

Here e the dimensionless parameter $\nu = d^{2}\nu\beta/\varepsilon_{0}$ describes the ratio of the electric interaction energy of two adjacent dipoles to thermal energy, $\hat{n}$ is the tensor which components are $n_{ij} = n_i\, n_j$ **.** Using the explicit T -matrix components we obtain explicit expressions for a set of effective dielectric constants which occur in the free energy.

To find the director profile it is necessary to minimize the corresponding free energy of the LC cell to obtain the differential equations for director with appropriate boundary conditions and to solve them.

The total free energy functional of the LC sample is defined by

$$F = F_{elastic} + F_{h\nu} + F_{E} + F_{flex} + F_{p} + F_{S}\,, \tag{12}$$

where the bulk elastic energy $F_{elastic}$ has a form

$$F_{elastic} = \frac{1}{2}K_{11}\int\left(\nabla\cdot\mathbf{n}\right)^{2}dV + \frac{1}{2}K_{22}\int\left(\mathbf{n}\cdot\nabla\times\mathbf{n}\right)^{2}dV + \frac{1}{2}K_{33}\int\left(\mathbf{n}\times\nabla\times\mathbf{n}\right)^{2}dV \tag{13}$$

Here $\mathbf{n}$ is the LC director, K_{11}, K_{22}, K_{33} are the splay, twist and bend elastic constants respectively.

The dc-(or ac-) electric fields contribution to the thermodynamic potential is as follows $F_{E} = -\frac{1}{8\pi}\varepsilon_{a}\int\left(\mathbf{n}\cdot\mathbf{E}\right)^{2}dV$, where $\varepsilon_{a} = \varepsilon_{\parallel} - \varepsilon_{\perp}$ is the anisotropy of the LC dielectric tensor, it may be positive or negative, depending on the chemical structure of LC molecules.

Flexoelectric polarisation gives rise to the following contribution in the total free energy $F_{flex} = -\int\left(\mathbf{P}_{f}\cdot\mathbf{E}\right)dV$, where $\mathbf{P}_{f} = e_{11}\mathbf{n}\nabla\cdot\mathbf{n} + e_{33}(\nabla\times\mathbf{n}\times\mathbf{n})$ and e_{11}, e_{33} are flexoelectric moduli.

F_{p} describes interaction of particles with external field. In the case of ferroelectric particles which create the electric polarization $\mathbf{P}_{p}$ this term has a form $F_{p} = -\int\left(\mathbf{P}_{p}\cdot\mathbf{E}\right)dV$. The analogous expression one can write for the case of ferromagnetic particles in external magnetic field.

The surface term F_{S} in the total thermodynamic potential originates from two contributions: LC-cell walls interaction and interaction between the particles and LC. A simple and very popular expression for F_{S} reads:

$$F_S = -\frac{1}{2} W \int \left(\mathbf{n} \cdot \mathbf{e} \right)^2 dS \qquad (14)$$

Here easy axis $\mathbf{e}$ is the averaged nematic director orientation on the surface minimising the surface part F_S of the total elastic energy in the absence of the bulk distortion. Light field contribution to the total free energy functional has the form

$$F_{light} = \frac{\tilde{\varepsilon}_a}{16\pi} \int \left(\mathbf{n} \cdot \mathbf{E}_{h\nu} \right)^2 dV \qquad (15)$$

where $\tilde{\varepsilon}_a$ is the anisotropy of the LC dielectric tensor at optical frequency.

Assuming there are no free charges the equation for the electric field in the nematic LC obeys the equation

$$div(\hat{\varepsilon} \cdot \mathbf{E} + \mathbf{P}_f + \mathbf{P}_p) = 0 \qquad (16)$$

where $\varepsilon_{ij} = \varepsilon_{\perp} \delta_{ij} + \varepsilon_a n_i n_j$ is the low frequency dielectric permittivity of LC, n_i are the components of director **n.** Equations for the electric field and the director are coupled and must be solved simultaneously.

The propagation of light in non-linear LC medium is governed by the Maxwell's equationis with the nonlinear polarisation $\mathbf{P}_{h\nu}^{NL}$ of the form $\mathbf{P}_{h\nu}^{NL} = \left(\tilde{\varepsilon}\left(\mathbf{r}\right) - \hat{I} \right) \cdot \mathbf{E}_{h\nu}$ where dielectric tensor at optical frequency

$$\tilde{\varepsilon}_{ij}\left(\mathbf{r}\right) = \tilde{\varepsilon}_{\perp} \delta_{ij} + \tilde{\varepsilon}_a \mathbf{n}_i \left(\mathbf{r}, \mathbf{E}_{h\nu}\right) \mathbf{n}_j \left(\mathbf{r}, \mathbf{E}_{h\nu}\right) \qquad (17)$$

We shall use approximations of a plane wave and slowly varying envelope to get the coupled-wave equations resulting from the index gratings. Two-wave coupled wave equations will be solved (both analytically and numerically) for the cases of the nematic LC doped with ferroic nano-particles. Then diffraction efficiency and two-beam coupling gain in the hybrid LC cells with ferroic nano-particles can be calculated.

Technical progress overview

Equation for polarization of isotropic liquid host filled with ferroelectric nano-particles is obtained on the base of the Fokker-Planck equation for orientational distribution function in the external electric field (AC/DC). Solution of equation for polarization gives possibility to calculate the displacement current i_d in the system for different concentrations of nano-particles and different repetition rates of external electric field. Theoretical results for the displacement current versus applied voltage describe well the experimental curves $i_d(U)$ using three or four fitting parameters for each species of suspension depending on the nano-particles concentration. Fitting theoretical curves for the displacement current versus external voltage with experimental curves allowed us to make suppositions on form and structure of nanoparticle compositions in suspension at different repetition rates of external voltage. We believe that one or two kinds of nanoparticle compositions in the form of dual nanoparticles and chain of nanoparticles together with single nanoparticles prevail in 4.5 wt. % suspension of 9 nm $BaTiO_3$ ferroelectric nanoparticles. We estimated values of electric dipoles, rotational relaxation times, rotational viscosity coefficients and volume concentrations of nanoparticles species in suspension in dependence on the voltage repetition rate.

We developed a theoretical model of the dielectric properties of a ferroelectric LC suspension, using a generalized Maxwell-Garnett theory. The suspension consists of an anisotropic matrix with a low concentration of impurity ferroelectric nanoparticles. The impurity particles possess shape and dielectric anisotropy, as well as a permanent electric polarization and strong LC director anchoring on the particle surface. We show that there are two sets of the effective permittivity tensors. One of these can be used to derive the dielectric properties like capacitances. A second set must be used in free-energy calculations to predict director profile in the cells where, for example, director field is inhomogeneous. Neglecting positional correlations of the nanoparticles we get expressions for both sets of the effective permittivity tensors.

Considering as an example Freedericksz transition in a ferroelectric LC suspension we developed a theoretical model which shows that spatially inhomogeneous distribution of impurity ferroelectric nanoparticles in a LC suspension can be very important for dielectric and optical properties of suspension. We have shown that under condition when inhomogeneous distribution of impurity ferroelectric nanoparticles is stabilized Freedericksz transition threshold can change its value at change of applied voltage sign. We found dependence of these Freedericksz transition thresholds and their asymmetry on spontaneous polarization of nanoparticles, weight concentration of nanoparticles, pattern of their spatial distribution and director anchoring energy on the cell substrates. In particular, we have shown that there is an area of parameters values, close to the experimental data, which well describes experimental results for Freedericksz transition threshold in the TL205 LC cell doped with $BaTiO_3$ ferroelectric nanoparticles.

We considered different cases of collective behavior of ferroelectric nanoparticles in the LC cell and studied their influence on electric field penetrating into the cell from photorefractive substrates. For these cases we calculated spontaneous polarization which nanoparticles create and took it into account to determine electric field in the cell. The analytical expressions for electric field are obtained. We show that in these expressions the spontaneous polarization of nanoparticles renormalizes the principal components of dielectric function and diffraction grating wave number depending on the type of nanoparticle collective behavior.

Dielectric function of the ferroelectric nanoparticles suspension in nematic LC cell with photorefractive substrates is investigated. Influence of electric field penetrating into the cell from photorefractive substrates on dielectric function of suspension is studied. We found that dielectric function of suspension depends essentially on director anchoring on the nanoparticle surface. For the cases of planar and homeotropic director anchoring we obtained analytical expressions which allow to calculate dielectric function at different values of ferroelectric nanoparticle suspension parameters.

We studied the impact of ferroelectric nano-particles on two-beam coupling gain in the hybrid nematic LC cells. We established that model of strong correlation of nano-particles orientation with the LC director field is applicable for describing experimental results for gain coefficient in LC TL205 doped by $BaTiO_3$ nano-particles. In this model gain coefficient changes its sign and becomes negative with increase of grating spacing and nanoparticle concentration. Gain coefficient depends also on mutual orientation of nano-particles long axis and LC director as well as a sign of dielectric anisotropy of ferroelectric suspension on light frequency.

Current status of the project.

Current technical status of the project: on schedule.

Summary of personnel commitment.

Professors of Kyiv National Taras Shevchenko University V. Yu. Reshetnyak and I. P. Pinkevych fulfilled the tasks during the reported period.

At the first stage of the project.

V.Yu. Reshetnyak – has solved the Fokker-Planck equation for the ferroelectric nano-particles re-orientation in the isotropic host under the externally applied electric field (AC/DC),

- has solved the dynamic equation for re-orientation of ferroelectric nano-particles in the isotropic suspension under the external electric field including inertia term, viscosity term ,
- has developed a generalized Maxwell-Garnett type theory of the low concentrated ferroic LC nano-suspension for the effective dielectric function,
- has studied the impact of director spatial inhomogeneity on the dielectric/magnetic response of the LC suspensions at the infinitely rigid director anchoring.

I.P. Pinkevych - has calculated the displacement current and studied the impact of concentration and chaining of the ferroelectric nano-particles on the dielectric response of the isotropic suspension,

- has solved the dynamic equation for re-orientation of ferroelectric nano-particles in the isotropic suspension under the external electric field including random forces;
- has developed a generalized Maxwell-Garnett type theory of the low concentrated ferroic LC nano-suspension for the effective magnetic function,
- has studied the impact of director spatial inhomogeneity on the dielectric/magnetic response of the LC suspensions at the finite director anchoring.

At the second stage of the project.
Prof. V. Reshetnyak has studied the impact of particle concentration inhomogeneity on the dielectric/magnetic response of the LC suspensions at the infinitely rigid director anchoring;
- the conditions for individual and collective behavior (response) of nano-particles of different nature in the suspensions at the infinitely rigid director anchoring;
- the influence of the tilt angle at the particles surface on the effective dielectric/magnetic function of suspension;.
- the impact of ferroic nano-particles on diffraction efficiency in the hybrid LC cells.

Prof. I. Pinkevych has studied the impact of particle concentration inhomogeneity on the dielectric/magnetic response of the LC suspensions at the finite director anchoring;
- the conditions for individual and collective behavior (response) of nano-particles of different nature in the suspensions at the finite director anchoring;
- the influence of the anchoring energy at the particles surface on the effective dielectric/magnetic function of suspension;
- the impact of ferroic nano-particles on two-beam coupling gain in the hybrid LC cells. It forms 50% of all tasks planned for period under report.

Performed by V.Yu. Reshetnyak investigations form 50% of all tasks planned for the period under report
Performed by I.P. Pinkevych investigations form 50% of all tasks planned for the period under report.

Variations in the scheduled amounts of efforts were absent.

Description of travels.

There were travels of prof. V.Yu.Reshetnyak:
to Yerevan, Armenia for participation in 14th International Topical Meeting on Optics of Liquid Crystals 2011, September 25 - October 1, 2011. At the conference V.Yu. Reshetnyak delivered two reports on scientific results of the project.

to Mainz, Germany, for participation in conference ILCC’12 (24-th International Liquid Crystal Conference), August 19 - August 26, 2012. Two talks on results of the project were given at the conference.

to Hong Kong, for participation in the 4th Workshop on Liquid Crystals for Photonics, and to Japan, Fuji Calm, for participation in the 1st Asian Conference on Liquid Crystals, December 7 - December 20, 2012. Two talks on results of the project were given at these conferences.

There was a travel of prof. I. Pinkevych to Cambridge, United Kingdom (May 16 – June 3, 2013) to participate in the programme “The Mathematics of Liquid Crystals” in the Isaac Newton Institute for Mathematical Sciences. One talk on results of the project was given.

Information about major equipment and materials acquired, other direct costs, related to the project.

During the reported period two notebooks and materials were purchased.

Table Redirection

Reference documents &date (1)	New requested category, or old category with new cost (2)	Requested cost (new) (3)	Original (old) category (4)	Estimated cost (old) (5)	Redirected cost (6) old – new
Quarter 9					
L02	Resource, 20275	2,025.63	5220 Project Exp-Travel Outside CR	5,943.31	2,940.80
L02	Resource, 20276	2,025.63	5210 6.6 ODC	779.09	779.09
L02	5205 5.4 Materials	386.40	I606116 Notebook, 606119 Notebook	717.77	717.77
Total by L02					4437.66