Optical control of graphene plasmon using liquid crystal layer 29K New One

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SCIENCE AND TECHNOLOGY CENTER IN UKRAINE

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Final Report

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The project is devoted to the basic research and establishes possible optical ways to control the surface plasmon polariton in graphene layer. A system comprises the graphene ribbons grating placed between a nematic liquid crystal (LC) and an isotropic dielectric. Incident light wave excites a plasmon in the graphene ribbons, which influences the light propagation in the system. The grating structure of the graphene monolayer is necessary to provide the plasmon excitation because there is a huge wave vector mismatch between the graphene plasmonic wave and the incident electromagnetic wave. In graphene structures the resonant plasmon frequency depends on the dielectric properties of layers placed above and below the graphene sheet or ribbon. In our case, it means that plasmon frequency must depend on the LC dielectric permittivity. The LC dielectric permittivity depends on the LC director orientational state, which can be controlled by external field. Therefore, a propagation of the surface plasmon polariton in graphene layer can be tuned by reorienting the LC director using external electric or magnetic fields.

14. SUBJECT TERMS
novel organic-inorganic structures, graphene and liquid crystal layers., plasmonic waves in graphene, EOARD, STCU
<table>
<thead>
<tr>
<th>Short form</th>
<th>File/page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Executive summary</td>
<td>SF.doc PAGE 1</td>
</tr>
<tr>
<td>Cooperation with foreign collaborators</td>
<td>SF.doc PAGE 2</td>
</tr>
<tr>
<td>Publications</td>
<td>SF.doc PAGE 2</td>
</tr>
<tr>
<td>Prospects of future development   (for final report only)</td>
<td>SF.doc PAGE 2</td>
</tr>
<tr>
<td>Full form</td>
<td></td>
</tr>
<tr>
<td>Project main idea</td>
<td>FF.doc PAGE 2</td>
</tr>
<tr>
<td>Technical approach</td>
<td>FF.doc PAGE 2</td>
</tr>
<tr>
<td>Technical progress overview</td>
<td>FF.doc PAGE 7</td>
</tr>
<tr>
<td>Current status of the project</td>
<td>FF.doc PAGE 10</td>
</tr>
<tr>
<td>Summary of personnel commitment</td>
<td>FF.doc PAGE 10</td>
</tr>
<tr>
<td>Description of travels</td>
<td>FF.doc PAGE 10</td>
</tr>
<tr>
<td>Information about major equipment and materials acquired, other direct costs, related to the project</td>
<td>FF.doc PAGE 10</td>
</tr>
</tbody>
</table>

1. Expression for plasmon frequency in the hybrid system with the LC layer.

4. Results and Discussion  
   T1.2.doc PAGE 3

2. Plasmon frequency dependence on the orientational state of the LC layer.

4. Results and Discussion  
   T1.2.doc PAGE 3

3. Expressions for coefficients of light reflection and transmission in the hybrid system.

4. Results and Discussion  
   T2.1.doc PAGE 4

4. Optimal parameters for active control of light propagation and reflection.

4. Results and Discussion  
   T2.2.doc PAGE 3
Project main idea

The project is devoted to the basic research and establishes possible optical ways to control the surface plasmon polariton in graphene layer. A system comprises the graphene ribbons grating placed between a nematic liquid crystal (LC) and an isotropic dielectric. Incident light wave excites a plasmon in the graphene ribbons, which influences the light propagation in the system. The grating structure of the graphene monolayer is necessary to provide the plasmon excitation because there is a huge wave vector mismatch between the graphene plasmonic wave and the incident electromagnetic wave. In graphene structures the resonant plasmon frequency depends on the dielectric properties of layers placed above and below the graphene sheet or ribbon. In our case, it means that plasmon frequency must depend on the LC dielectric permittivity. The LC dielectric permittivity depends on the LC director orientational state, which can be controlled by external field. Therefore, a propagation of the surface plasmon polariton in graphene layer can be tuned by reorienting the LC director using external electric or magnetic fields.

After establishing the plasmon frequency dependence on the LC director orientational state we shall study a light wave propagation across the system under consideration. We shall calculate the absorption, reflection, and transmission coefficients and show that control of the orientational state of the LC layer enables us to manipulate by magnitude of the absorption/reflection maxima.

These results will allow suggesting a new type of the graphene micro-ribbon grating structures to control light beams propagation.

Technical approach

Dependence of plasmon frequency on the LC director orientational state.

Schematic of the hybrid system, LC – graphene monolayer – isotropic dielectric, is presented in Fig. 1.

![Schematic of the hybrid system](image)

Fig. 1. Schematic of the hybrid system: LC – graphene monolayer – isotropic dielectric.

We use the following designations: \( \hat{\varepsilon}_1 \) is the LC dielectric tensor, \( \varepsilon_2 \) is the isotropic dielectric permittivity, \( \sigma \) is the graphene surface conductivity, \( \psi, \varphi \) are the LC director angles.

We write the Maxwell equations for system shown in Fig. 1 and seek their solution in the following form:

1) for the wave in the LC \( (z > 0) \)

\[
E_1 = (e_x E_{1x} + e_z E_{1z}) e^{i(q_x-\omega t)z}, \quad H_1 = e_y H_{1y} e^{i(q_x-\omega t)z},
\]

2) for the wave in the isotropic dielectric \( (z < 0) \)

\[
E_2 = (e_x E_{2x} + e_z E_{2z}) e^{i(q_x-\omega t)+Q_2 z}, \quad H_2 = e_y H_{2y} e^{i(q_x-\omega t)+Q_2 z}.
\]
Substituting (2), (3) into Maxwell's equations we take into account that the LC dielectric tensor has a general form

\[
\hat{\epsilon}_i = \begin{pmatrix}
\epsilon_{xx} & \epsilon_{xy} & \epsilon_{xz} \\
\epsilon_{xy} & \epsilon_{yy} & \epsilon_{yz} \\
\epsilon_{xz} & \epsilon_{yz} & \epsilon_{zz}
\end{pmatrix},
\]

(3)

where on optical frequencies

\[
\epsilon_{ij} = n_o^2 \delta_{ij} + \left(n_e^2 - n_o^2 \right) d_i d_j.
\]

(4)

Here \(d_i\) is the director Cartesian component, \(n_o, n_e\) are the refractive indices for ordinary and extraordinary waves in the LC, respectively.

To the expressions obtained after substitution (1), (2) into Maxwell's equations we must add the boundary conditions,

\[
E_z|_{z=0} = E_{ix}|_{z=0}, \quad \left(H_{2y} - H_{1y}\right)|_{z=0} = \sigma E_{ix}|_{z=0}
\]

(5)

Demanding the boundary conditions (5) to be satisfied we obtain after some algebraic transformations a dispersion equation for plasmon wave propagating in the graphene

\[
\sqrt{\frac{\epsilon_{xx}\epsilon_{zz} - \epsilon_{xz}^2}{q^2 - \omega^2/c^2}} - \frac{\epsilon_2}{\sqrt{q^2 - \epsilon_2^2 / c^2}} = -\frac{i \sigma}{\omega \epsilon_0},
\]

(6)

were

\[
\begin{align*}
\epsilon_{xx} &= n_o^2 + \left(n_e^2 - n_o^2 \right) \cos^2 \varphi \sin^2 \psi, \\
\epsilon_{zz} &= n_o^2 + \left(n_e^2 - n_o^2 \right) \cos^2 \psi, \\
\epsilon_{xz} &= \left(n_e^2 - n_o^2 \right) \cos \varphi \sin \psi \cos \psi
\end{align*}
\]

(7)

and the LC director angles are shown in Fig. 1.

The graphene surface conductivity \(\sigma\) in (6), in general case, can be expressed as a sum of two terms, \(\sigma = \sigma_{\text{int,ra}} + \sigma_{\text{int,er}}\), where the first term corresponds to the intraband electron transitions and the second term gives contribution from the interband transitions. For \(\hbar \omega >> k_B T\) and for the Fermi energy \(|E_F| >> k_B T\) it can be approximately written as [1],

\[
\begin{align*}
\sigma_{\text{int,ra}} &= \frac{i e^2 |E_F|}{\pi \hbar^2 (\omega + i / \tau)}, \\
\sigma_{\text{int,er}} &= \frac{i e^2}{4 \pi \hbar} \ln \left( \frac{2 |E_F| - \hbar (\omega + i / \tau)}{2 |E_F| + \hbar (\omega + i / \tau)} \right)
\end{align*}
\]

(8)

In the nonretarded limit \(q >> \omega \epsilon / c\) the dispersion equation (6) simplifies and we arrive at expression

\[
q = \frac{i \omega \epsilon_0}{\sigma} \sqrt{\frac{\epsilon_{xx} \epsilon_{zz} - \epsilon_{xz}^2}{\epsilon_2}},
\]

(9)

It is seen that influence of the LC orientational state on the dispersion equation is described by term
Reflection and transmission coefficients.

Consider a graphene micro-ribbon grating in the xy-plane placed between a nematic LC layer (anisotropic top substrate) and an isotropic dielectric substrate. Each micro-ribbon in the grating is a single layer graphene with the ribbon along the y-axis, where d is the micro-ribbon width and \( \Lambda \) is the grating spacing (Fig. 2). A plane monochromatic wave propagates along the z-axis from the side of the LC and excites the plasmons in the graphene micro-ribbons. We assume semi-infinite substrates that allows us to neglect the effects of multiple reflections; in the y-direction the system is infinite.

To simplify calculations we suppose the LC director to be only reoriented in the xz-plane, where the angle \( \psi \) describes the director deviation from the z-axis. We also set the magnetic vector of the incident wave to be perpendicular to the xz-plane (TM-wave).

\[
E(\varphi, \psi) = \frac{e_{xx}e_{zz} - e_{xz}^2}{n_e n_o} \sin^2 \varphi \cos^2 \psi + \cos^2 \varphi + \left(\frac{n_o}{n_e}\right)^2 \sin^2 \varphi \sin^2 \psi
\]  

(10)

Fig. 2. Schematic of the graphene micro-ribbon grating structure. d is the ribbon width, \( \Lambda \) is the grating spacing, \( \psi \) is the director angle with the z-axis, \( \hat{e}_1 \) is the LC dielectric tensor, \( \varepsilon_2 \) is the isotropic substrate dielectric permittivity, \( \sigma \) is the graphene conductivity, \( e_n \) is a normal to the graphene plane, and \( k_i \) is the wave vector of an incident plane monochromatic wave.

Because of the strong confinement of the surface plasmon-polaritons (SPPs) in the ribbons, only the thin layer of the LC substrate near the graphene influences the SPPs. Therefore, we can set the LC director orientation in the whole substrate to be homogeneous and equal to the director orientation of this layer near the graphene. The LC optical dielectric tensor can be written in the form \( \varepsilon_{ij} = n_i^2 \delta_{ij} + (n_o^2 - n_i^2)n_i n_j \), where \( n_i \) denotes the components of the director \( n = (\sin \psi, 0, \cos \psi) \); \( n_o \) and \( n_e \) are the refractive indices of the ordinary and extraordinary waves, respectively [2].

We suppose that the TM-wave is normally incident in the LC on the graphene grating. As it follows from the Maxwell equations, electric and magnetic vectors of this wave in the LC take the form:

\[
\mathbf{E}_i = (E_x \mathbf{e}_x + E_z \mathbf{e}_z) e^{-i(k_i z + \omega t)}, \quad \mathbf{H}_i = -\frac{k_i}{\omega \mu_0} E_{ix} e^{-i(k_i z + \omega t)} \mathbf{e}_y, \quad E_{iz} = -\varepsilon_{1zz} / \varepsilon_{1xx} E_{ix},
\]  

(11)

with the dispersion equation

\[
k_i = \frac{\omega}{c} \sqrt{\frac{\varepsilon_{1xx}}{\varepsilon_{1zz}}}. 
\]  

(12)
For reflected and transmitted waves, we use the Fourier-Floquet expansion with respect to the coordinate $x$. Satisfying the Maxwell equations, the electric and magnetic vectors of these waves can be written as follows:

i) for the reflected wave

$$
E_r = \frac{c}{\sqrt{\epsilon_{1xx} - \epsilon_{1zz}^2 / \epsilon_{1zz}}} \sum_n \left[ (\epsilon_{1xx} k_m + \epsilon_{1zz} k_n) \mathbf{e}_x - (\epsilon_{1xx} k_m + \epsilon_{1zz} k_n) \mathbf{e}_z \right] a_n e^{i(k_x x + k_n z - \omega t)},
$$

$$
H_r = \epsilon_0 c \sqrt{\epsilon_{1xx} - \epsilon_{1zz}^2 / \epsilon_{1zz}} \sum_n a_n e^{i(k_x x + k_n z - \omega t)} \mathbf{e}_y,
$$

with the dispersion equation

$$
k_m = \sqrt{\left(\epsilon_{1xx} - \epsilon_{1zz}^2 / \epsilon_{1zz}\right) - \frac{\omega^2}{c^2} - \epsilon_{1zz} k_n},
$$

(14)

ii) for the transmitted wave

$$
E_t = -\frac{c}{\omega \sqrt{\epsilon_2}} \sum_n (k_{nt} \mathbf{e}_x + k_{nt} \mathbf{e}_z) b_n e^{i(k_x x - k_n z - \omega t)},
$$

$$
H_t = \epsilon_0 c \sqrt{\epsilon_2} \sum_n b_n e^{i(k_x x - k_n z - \omega t)} \mathbf{e}_y,
$$

with the dispersion equation

$$
k_n = \sqrt{\frac{\omega^2}{c^2} - \epsilon_2 - k_n^2}.
$$

(16)

Here $k_n = 2\pi n / \Lambda$, $n$ is the number of a Floquet spatial harmonic.

We define the current density in the plane of the graphene grating as $j_x = \sigma(x) E_{tx} (z = 0) \mathbf{e}_x$, where $\sigma(x) = \sigma$ within each micro-ribbon and $\sigma(x) = 0$ in the gaps between micro-ribbons. Substituting $E_{tx} (z = 0)$ from eq. (15), we obtain

$$
j_x = -\sigma(x) \frac{c}{\omega \sqrt{\epsilon_2}} \sum_n k_{nt} b_n e^{i(k_x x - \omega t)} \mathbf{e}_x.
$$

(17)

To obtain equations for coefficients $a_n, b_n$ of the Fourier-Floquet expansions (13), (15) we use the boundary conditions:

$$
\left[ \mathbf{e}_n \times (\mathbf{H}_t + \mathbf{H}_r - \mathbf{H}_t) - j_x \right]_{z=0} \cdot \mathbf{e}_x = 0,
$$

$$
\left[ \mathbf{e}_n \times (\mathbf{E}_t + \mathbf{E}_r - \mathbf{E}_t) \right]_{z=0} \cdot \mathbf{e}_y = 0.
$$

(18)

Now we substitute eqs. (11), (13), (15) for electric and magnetic vectors and eq.(17) for the current density $j_x$ into eqs. (18). Then, using the dispersion equations (12), (14), (16) we arrive at
\[ \sum_n \left[ \frac{c_{1xx}^2 - c_{1zz}^2}{c_{1zz}} a_n - \left( \frac{e^2}{c^2} + \frac{4 \pi^2 n^2}{e^2 \Lambda^2} \sigma(x) \right) b_n \right] \frac{2 i \pi n x}{\Lambda} - \sqrt{c_{1xx}^2 - c_{1zz}^2} E_{1s} = 0 \]  
(19)

\[ \sum_n \left[ \frac{\omega^2 - 4 \pi^2 n^2}{c^2 - \frac{e^2 \Lambda^2}{e^2 \Lambda^2}} a_n + \frac{\omega^2 - 4 \pi^2 n^2}{c^2 - \frac{e^2 \Lambda^2}{e^2 \Lambda^2}} b_n \right] \frac{2 i \pi n x}{\Lambda} + \frac{\omega}{c} E_{1s} = 0 \]  
(20)

Multiplying eqs. (19), (20) by \( e^{-2 i \pi m x / \Lambda} \), where \( m \) is an integer and integrating these equations over \( x \) in the range \([0, \Lambda]\) we obtain the set of equations for coefficients \( a_n, b_n \):

\[ b_n = \frac{1}{\sqrt{c^2 - \frac{e^2 \Lambda^2}{e^2 \Lambda^2}}} \left( \frac{\omega}{c} E_{1s} \delta_{m0} - \sqrt{\frac{\omega^2 - 4 \pi^2 n^2}{c^2 - \frac{e^2 \Lambda^2}{e^2 \Lambda^2}}} a_n \right), \]

\[ \sum_{m(n \neq 0)} \frac{i \sigma \left( e^{-2 i \pi n d / \Lambda} - 1 \right)}{2 \pi \omega e_0 (n-m)} a_m = \]

\[ \left[ \frac{\omega^2 - 4 \pi^2 n^2}{c^2 - \frac{e^2 \Lambda^2}{e^2 \Lambda^2}} - \frac{\sigma d}{e_0 \Lambda} \right] \frac{\sqrt{c^2 - \frac{e^2 \Lambda^2}{e^2 \Lambda^2}}}{e_{1xx}^2 - e_{1zz}^2} a_n + \frac{i \sigma \left( 1 - e^{-2 i \pi n d / \Lambda} \right)}{2 \pi e_0 n c} E_{1s} \delta_{n0} + \frac{\sqrt{c^2 - \frac{e^2 \Lambda^2}{e^2 \Lambda^2}}}{e_{1xx}^2 - e_{1zz}^2} a_n \]

In the infrared and terahertz spectral regions the intraband contribution to the graphene conductivity dominates. Then, in the random phase approximation the 2D conductivity of graphene can be written as [3]

\[ \sigma = \frac{e^2}{\pi \hbar^2} \frac{\tau i}{\omega \tau + i}, \]

(23)

where \( E_F \) is the Fermi energy of graphene, \( \tau \) is the electron relaxation time, \( e \) is the electron charge.

The reflection and transmission coefficients are defined as

\[ R = \left| \text{Re}(E_x \times H_y') / \text{Re}(E_y \times H_x') \right|, \quad T = \left| \text{Re}(E_y \times H_y') / \text{Re}(E_x \times H_x') \right|. \]

(24)
The graphene absorption coefficient is \( A = 1 - (R + T) \). Numerically solving equations (21)-(23) the coefficients \( a_n, b_n \) are obtained. Using formulas (13), (15) to find the electric and magnetic vectors of the reflected and transmitted waves and after their substitution into formula (24), we calculate the reflection, transmission, and absorption coefficients.

**Technical progress overview**

In Fig. 3 we show dependence of quantity \( E(\phi, \psi) \) on the LC director angles \( \phi, \psi \) for two pairs of the refractive indices of the ordinary and extraordinary waves in the LC, \( n_o, n_e \).

![Graphical representation of E(\phi, \psi) for two pairs of refractive indices.](image)

**Fig. 3.** \( E(\phi, \psi) \) versus \( \phi, \psi \): (a) \( n_o = 1.53, n_e = 1.94 \); (b) \( n_o = 1.94, n_e = 1.53 \).

Below in Fig. 4 we also show for illustration the cross-sections of function \( E(\phi, \psi) \) by vertical planes.

![Graphical representation of cross-sections of E(\phi, \psi) for two pairs of refractive indices.](image)

**Fig. 4.** Cross-sections of function \( E(\phi, \psi) \) by vertical planes: (a) \( n_o = 1.53, n_e = 1.94 \); (b) \( n_o = 1.94, n_e = 1.53 \).
Analyzing formula (10) (or Figs. 3, 4) we can conclude that the ratio \( E_{\text{max}} / E_{\text{min}} \) responsible for interval of tuning the plasmon wave vector (or frequency) by the LC director reorientation equals \( n_e / n_o \) if \( n_e > n_o \) or \( n_o / n_e \) if \( n_o > n_e \). It is necessary to note that in the retarded region (\( q < \omega \varepsilon_{\infty} / c \)) where instead of formula (9) we have to use formula (6), the small correction to this estimation can arise from \( \varepsilon_{zz} \) in denominator of formula (6).

As material of the nematic layer, we use the high birefringence (\( \Delta n \approx 0.41 \)) LC mixture W1791 with \( n_o \approx 1.53, n_e \approx 1.94 \) at \( \lambda = 1.064 \mu m \) [4]. The birefringence of W1791, which is known in visible and near-IR regions of spectrum, was extrapolated to mid-IR and THz frequencies using the extended Cauchy dispersion formulas obtained for the LC mixture E7 [5]. In our study, we chose either a low dielectric constant material (e.g. hexagonal boron nitride, h-BN, \( \varepsilon_2 = \varepsilon_{h-BN} = 3 \) ) [6, 7] or a high dielectric constant material (silicon, Si, \( \varepsilon_2 = \varepsilon_{Si} = 11.7 \) ) [8] as the isotropic substrate material.

The physical parameters of the graphene grating in our simulations are as follows: the grating spacing was fixed at \( \Lambda = 1 \mu m \), the micro-ribbon width was varied as \( d = 0.3 \Lambda, 0.5 \Lambda \) and \( 0.7 \Lambda \). For evaluation of the electron relaxation time we used the formula \( \tau = \mu E_{F} / (e \varepsilon_{F}) \), where \( \mu \) is the carrier mobility, \( \varepsilon_{F} = 3 \times 10^6 \text{m} \cdot \text{s}^{-1} \) was the Fermi velocity in graphene [10]. Setting \( E_{F} = 0.64 \text{ eV} \) [9] and the carrier mobility \( \mu = 0.5 \ \text{m}^2/(\text{V} \cdot \text{s}) \) we get the carrier scattering time \( \tau = 0.32 \text{ ps} \). The simulations were performed in the wavelength range 14.5-55 \( \mu m \) (20-5.45 THz) with a temperature \( T = 300 \text{ K} \).

The convergence of the computational procedure used with respect to higher Fouquet harmonics is necessary. In order to ensure the required accuracy of calculations, it was sufficient to select the number of harmonics \( N \geq 400 \) in eqs. (21), (22). Results of our numerical calculations of absorption, reflection, and transmission spectra of the graphene grating are shown in Figs. 5 and 6. Fig. 5 illustrates the change of the absorption, reflection, and transmission spectra of the graphene grating for two limiting values of the LC director angle \( \psi = 0^\circ \) and \( \psi = 90^\circ \), and a ribbon width to grating spacing ratio \( d / \Lambda = 0.5 \). Curves 1 and 2 correspond to the cases of an isotropic substrate with either a low or a high dielectric constant, respectively. Maxima in the absorption and reflection spectra are related to the excitation of the plasmons in the graphene micro-ribbons. Within our frequency range two plasmon peaks in curves 2 are observed. The ratio of the resonant frequencies corresponding to these peaks is equal to 2, which agrees with the results obtained in paper [9] for the graphene grating placed between two isotropic dielectric substrates.

As we can see from Fig. 5, values of these maxima depend on the LC director orientation. In particular, the rotation of the LC director by \( 90^\circ \) leads to a change of the absorption maximum value by approximately 14% when \( \varepsilon_2 = 3 \) (h-BN substrate) and by 16% when \( \varepsilon_2 = 11.7 \) (Si substrate).
In Fig. 6, the change of the absorption, reflection, and transmission spectra of the graphene grating at the two limiting values of the LC director angle $\psi$ is shown for two values of the ribbon width to grating spacing ratio, $d / \Lambda = 0.3$ and $d / \Lambda = 0.7$. In this case, the rotation of the LC director by 90$^\circ$ leads to the change in the absorption by approximately 14% when $d / \Lambda = 0.3$ and by 18% when $d / \Lambda = 0.7$. A comparison of the results presented in Figs. 5, 6 shows that the effect of the LC director rotation increases with an increase of the isotropic substrate dielectric constant and the ribbon width to grating spacing ratio $d / \Lambda$.

On scientific results of the project one paper is published, one paper is prepared for publication, and four reports on international conferences were delivered.

References.
Current status of the project.
Current technical status of the project: on schedule.

Summary of personnel commitment.
Professors of Taras Shevchenko National University of Kyiv V. Yu. Reshetnyak, I. P. Pinkevych and Dr. V. I. Zadorozhni fulfilled the tasks during the reported period.

Prof. V. Yu. Reshetnyak - developed the mathematical model of the hybrid system with the LC layer,
- obtained expressions for plasmon frequency in the hybrid system with the LC layer,
- calculated the graphene plasmon frequency in dependence on the orientational state of the LC layer,
- obtained equations describing light propagation in the hybrid system with plasmon excitation in the graphene,
- developed numerical methods for calculation coefficients of light reflection and transmission in the hybrid system,
- optimized the hybrid system parameters for active control of light propagation and reflection.

Prof. I.P. Pinkevych - obtained and solved equations for the director and electric field in the LC cell,
- studied the influence of anchoring on the LC cell boundaries on the director in the cell bulk,
- studied the control of plasmon frequency by DC field and incident light field,
- obtained expressions for coefficients of light reflection and transmission in the hybrid system,
- studied influence of the LC layer orientational state on light reflection and transmission in the hybrid system.

Dr. V.I. Zadorozhni– fulfilled numerical calculations of light reflection and transmission coefficients in dependence on the anchoring conditions on the LC cell boundaries.

Variations in the scheduled amounts of efforts.
Prof. V. Yu. Reshetnyak and Prof. I.P. Pinkevych fulfilled the following tasks planned initially for Dr. V.I. Zadorozhni:
calculation of the graphene plasmon frequency in dependence on the orientational state of the LC layer (Reshetnyak),
develop numerical methods for calculation coefficients of light reflection and transmission in the hybrid system (Reshetnyak),
study the influence of anchoring on the LC cell boundaries on the director in the cell bulk (Pinkevych).

Description of travels.
Travel of prof. V. Yu. Reshetnyak for participation in Conference on Photorefractive Materials, Villars, Switzerland, 16-19 June 2015. Report was delivered on scientific results of the project.
Travels of prof. V. Yu. Reshetnyak and prof. I. P. Pinkevych for participation in 16th Topical Meeting on the Optics of Liquid Crystals, Sopot, Poland, 13-18 September, 2015. Report was delivered on scientific results of the project.

Information about major equipment and materials acquired, other direct costs, related to the project.
There was not purchase of equipment or materials, and other direct costs have not been spent.

Table Redirection

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