Light-induced Fredericks transition in the nematic liquid crystal cell with plasmonic nanoparticles at a cell bounding substrate

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The paper presents a theoretical description of the light-induced nematic liquid crystal reorientation in a cell with gold nanoparticles deposited on the surface of one of bounding substrates. It is shown that the surface plasmon resonance in the nanoparticles significantly affects the threshold of the director reorientation. The mathematical model of a surface free-energy density of nematic cell is given, which takes into account the influence of the local electric field on the near-surface nematic layer at the substrate with gold nanoparticles. The threshold intensity of a director orientation instability is calculated and its dependence on the wavelength of incident light and the degree of filling of the surface with gold nanoparticles is analyzed. Comparison of the theoretical calculations with experimental data confirms the full adequacy of the proposed theoretical model.

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I. INTRODUCTION

The main structural element of liquid crystal display technologies is a cell of liquid crystal (LC), in particular, nematic liquid crystal (NLC). Widespread practical application of LC is due to the phenomenon of orientational instability of a director in external electric and magnetic fields (Fredericks transition) [1]. In particular, a light-induced (i.e., optical) Fredericks transition (OFT) [2–5]—the threshold reorientation of the NLC director by an external light field is of great interest. These phenomena and prospects of their application have been considered in a number of works [6–19]. The addition of impurities of various materials, including nanomaterials, into the LC media has been promising on the way to improving the physical properties of liquid crystalline materials, and, accordingly, the functional characteristics of cells based on them. In particular, it was established in Ref. [20] that different impurities essentially differently affect the light-induced director reorientation even if their absorption coefficients are approximately equal. The study of dielectric properties of the suspension of NLC with ferroelectric colloidal particles was performed in Ref. [21]. The effect of a constant electric field on the giant optical nonlinearity of NLC doped with methyl red was studied in Ref. [22]. A decrease of the threshold voltage of the director orientational instability was observed when MgO [23] and Ti nanoparticles [24] of a small concentration were added into the NLC cell. The electrical Fredericks transition was studied both experimentally in the NLC cell doped with $BaTiO_3$ nanoparticles [25] and theoretically in the NLC suspension with nanosized ferroparticles [26]. As it was found in Refs. [27,28], the addition of gold nanoparticles into NLC media leads to a decrease of the threshold voltage and the nematic elastic constants, and to the increase of optical transmittance of the sample. In addition, the presence of gold nanoparticles in the bulk of NLC led to a decrease of the temperature of a nematic-isotropic liquid phase transition and an increase of the electrical conductivity of the system [29,30]. Light-induced changes of the refractive indices of nematic doped with gold nanoparticles were studied in Ref. [31]. The increases of the anisotropy of dielectric constant and rotational viscosity were observed in the NLC sample doped with ferroelectric nanoparticles [32–34]. Nonlinear optical effects in a NLC cell doped with gold nanoparticles were discussed in Refs. [35–39]. The authors of Ref. [40] proposed the theory of the effective medium, which is a LC with nanoscale inclusions of the "core-shell" type.

Despite the fact that the threshold reorientation of the director is a three-dimensional phenomenon, its nature and the threshold value significantly depend on the strength and type of interaction of LC with a cell surface. It is known that the effect of the cell surface can be significantly enough to provoke spontaneous or stimulated by altering surface conditions Fredericks transition [41–43]. One of the important parameters that determines conditions for the director at a surface is the anchoring energy. In Ref. [44] it was shown that the anchoring energy can be different when the director deviates from its easy axis of orientation in the azimuthal and polar directions. In Ref. [45], the light-induced change of surface conditions for the director in an azo dye-doped NLC cell was observed. The possibilities of improving the functional characteristics of LC cells are significantly expanded by the modification of the orienting surface with nanoparticles of different types. Thus, in the NLC cell with TiO_2 nanoparticles randomly deposited on one of the bounding surfaces, the increases of temperature sensitivity and electrical capacity of the system were observed [46]. The photothermal effect in a cell of cholesteric LC with gold nanoparticles covering one of the bounding surfaces and its impact on the optical properties of the sample were

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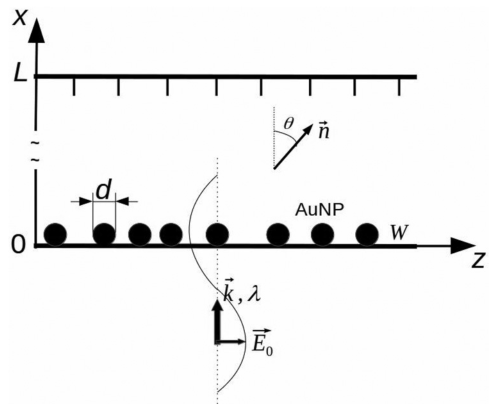

FIG. 1. Geometry of the problem.

studied in Ref. [47]. This effect was associated with plasmon resonance, which arises in the ensemble of gold nanoparticles in an incident light field. The overall reduction in power consumption of information display devices is directly related to lowering the threshold values. In this way, in Ref. [48] it was experimentally established that gold nanoparticles deposited on the substrate of a NLC cell make it possible to dynamically vary the effective anchoring energy of the nematic with the surface in a light wave field. Plasmon resonance induced by a light field in the ensemble of gold nanoparticles leads to a decrease in the threshold intensity of the OFT and the threshold magnitude of the magnetic field [48], when static properties of the orienting surface coating do not change.

In this study we present a theoretical description of the light-induced NLC director reorientation in a cell with gold nanoparticles deposited on the surface of one of bounding. We show that the surface plasmon resonance in the nanoparticles significantly affects the threshold in the director reorientation. The mathematical model of a surface free-energy density of NLC is proposed, which takes into account the influence of the local electric field on the director of the near-surface layer of NLC near the substrate with gold nanoparticles.

II. THEORETICAL MODEL

Let us consider a flat NLC cell bounded by the planes $x = 0$ and $x = L$ with initial homeotropic orientation of the director along the Ox axis. The initial orientation of the director is provided by the homeotropic alignment of NLC at the surface of the bounding substrates. We consider the anchoring of NLC with the surface $x = L$ to be infinitely strong, and the anchoring energy W on the surface $x = 0$ is finite. The gold nanoparticles are randomly deposited on the surface $x = 0$ of the lower substrate. The diameter d of the nanoparticles is of the order of several tens of nanometers [48]. A plane monochromatic light wave $\mathbf{E} = \mathbf{E}_0 \exp{(ikx - i\omega t)}$, polarized along the Oz axis, is incident normally on the NLC cell along Ox axis of the Cartesian coordinate system (see Fig. 1). The covering of the substrate with gold nanoparticles is considered to be noncontinuous, so that the light wave can penetrate the bulk of the cell almost without loss of intensity [48]. Also we do not take into account thermal effects that can be a consequence of light absorption. Such an assumption based on an experimental study [48] in which no heating of the sample was observed. Plasmon resonance occurs in the ensemble of gold nanoparticles under the action of incident light wave. This phenomenon implies a significant increase of the local electromagnetic field around each individual gold nanoparticle. Significantly enhanced local electric field, reorienting NLC molecules of the near-surface layer, disturbs the orienting effect of the substrate on which the gold nanoparticles are deposited.

The free energy of the NLC cell can be written in the form

$$\Phi = \Phi_{el} + \Phi_E + \Phi_S, \tag{1}$$

$$\Phi_{el} = \frac{1}{2}\int_V [K_1(\mathrm{div}\mathbf{n})^2 + K_2(\mathbf{n}\cdot\mathrm{rot}\mathbf{n})^2 + K_3[\mathbf{n}\times\mathrm{rot}\mathbf{n}]^2]\,dV,$$

$$\Phi_E = -\frac{1}{16\pi}\int_V \mathbf{E}\hat{\varepsilon}\mathbf{E}^*\,dV, \quad \Phi_S = \int_S F_S\,dS,$$

where Φ_{el} is an elastic energy of the NLC cell, Φ_E represents the contribution of the electric field of the light wave to the free energy, Φ_S and F_S denotes surface free energy and its density, respectively, K_1, K_2, and K_3 are the Frank's elastic constants, $\mathbf{n}$ is a director, $\hat{\varepsilon} = \varepsilon_\perp \hat{\mathbf{1}} + \varepsilon_a \vec{n}\otimes\vec{n}$, $\varepsilon_a = \varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$ are tensor and anisotropy of dielectric permeability of nematic at the frequency of incident light, $\mathbf{E}$ is an electric field of the light wave in the bulk of NLC cell, surface integral in Φ_S is taken over the surface $x = 0$ of the lower substrate.

Let us construct a model of the density F_S of the surface free energy of NLC. We shall suppose that the wavelength of the incident light is at least one order of magnitude larger than the linear size of the gold nanoparticles deposited on the surface of the substrate. Assuming that the filling of the substrate surface with gold nanoparticles is incomplete, the local electric field near the individual particle can be found in the quasi-static approximation [49,50]:

$$\mathbf{E}_{\mathrm{loc}} = \mathbf{E}^0_{\mathrm{loc}} \exp{(ikx - i\omega t)}, \tag{2}$$

where

$$\mathbf{E}^0_{\mathrm{loc}} = E_0\left[\left(1 + \frac{\alpha d^3}{4\tilde{r}^3}\right)\cos\tilde{\theta}\cdot\mathbf{e}_{\tilde{r}} + \left(-1 + \frac{\alpha d^3}{8\tilde{r}^3}\right)\sin\tilde{\theta}\cdot\mathbf{e}_{\tilde{\theta}}\right].$$

Here $\alpha = \frac{\varepsilon_1 - \varepsilon_m}{\varepsilon_1 + 2\varepsilon_m}$, $\varepsilon_m = \frac{\varepsilon_\parallel + 2\varepsilon_\perp}{3}$, ε_1 is a dielectric constant of gold, $\omega = \frac{2\pi c}{\lambda}$ is a frequency of the light wave in vacuum, $\mathbf{e}_{\tilde{r}}$, $\mathbf{e}_{\tilde{\theta}}$ denotes unit vectors of a spherical coordinate system starting at the center of the nanoparticle with the polar axis in the Oz direction, and $\tilde{r}$, $\tilde{\theta}$, $\tilde{\varphi}$ are spherical coordinates of a point with respect to the center of the nanoparticle.

The local electric field rapidly decreases as $1/\tilde{r}^3$ with distance from the center of the nanoparticle. Therefore, the influence of the local electric field of the ensemble of gold nanoparticles on the entire bulk of the NLC cell can be neglected. We consider that impact on the director of the NLC to be significant only in a narrow layer of thickness d near the surface of the lower cell substrate. Therefore, the contribution of the local electric field into the surface density F_S of the free energy of NLC can be expressed as follows:

$$F_S = -(1 - f_s)\frac{W}{2}(\mathbf{n}\mathbf{e})^2 - \gamma f_s \frac{\varepsilon_a}{8\pi}\langle(\mathbf{n}\mathbf{E}_{\mathrm{loc}})(\mathbf{n}\mathbf{E}^*_{\mathrm{loc}})\rangle_{S^*}\,d, \tag{3}$$

where f_s denotes a relative part of the surface occupied by gold nanoparticles, $\mathbf{e} = (1, 0, 0)$ is the unit vector describing the LC director orientation at cell substrate areas free from nanoparticles, $\gamma = \gamma(f_s)$ is a phenomenological coefficient that takes into account the local electric field amplification by the ensemble of nanoparticles near the surface of the bounding substrate and is calculated in accordance with the results of Refs. [51,52], averaging in the second term of F_S is performed over the surface S^* of a single nanoparticle [53]. The first term in F_S (3), written in the form of the Rapini potential, takes into account the anchoring of NLC with the surface area not filled with gold nanoparticles. The second term in F_S (3) takes into account the impact of the local electric field of the gold nanoparticles ensemble on the director of the NLC in the near-surface layer of the substrate occupied by nanoparticles. The anchoring of the NLC to the surface of gold nanoparticles is considered to be weak, so that the nanoparticles do not change the predominant homeotropic orientation of the director near the surface of the cell substrate in accordance with the description of the experiment in Ref. [48]. In the experimental work, after the deposition of gold nanoparticles, the surface of the cell substrate was covered with a surfactant to provide the predominant homeotropic orientation of the NLC on the entire surface. Thus, avoiding an explicit consideration of the orienting effect of individual gold nanoparticles on director orientation, the authors took into account the collective effect of the monolayer of gold nanoparticles, characterizing the anchoring of the NLC to the substrate both with and without nanoparticles by some effective value of the anchoring energy. Indeed, the presence of gold nanoparticles led to the decrease of the effective anchoring energy at most in twice [48]. The anchoring energy lowers as the time of nanoparticles deposition and, consequently, the covering degree f_s increase. However, according to our assessment, it cannot be the reason for such a significant decrease of the OFT threshold reported in Ref. [48]. In this case, the drop of the threshold value is due to the effect of the enhanced local electric field near the surface of an individual gold nanoparticle caused by plasmon resonance. The local electric field, averaged over the surface of a nanoparticle, turns out to be oriented parallel to the substrate along the incident light polarization, and, as a result, causes a significant weakening of the orienting action of the substrate surface, especially in the region near the deposited nanoparticles. In the proposed theoretical model within the plane deformations of the director field, we actually consider the collective influence of a monolayer of gold nanoparticles on the orientation of the NLC.

For the considered geometry of the initial homogeneous alignment $\mathbf{n}_0$ of the cell and the polarization $\mathbf{E}_0$ of the incident light wave, the threshold reorientation of the director is possible. We assume that the light-induced reorientation occurs in the plane xOz. Due to the homogeneity of the system in the Oy direction, a director in the bulk can be written in the form

$$\mathbf{n} = \cos\theta(x)\cdot\mathbf{i} + \sin\theta(x)\cdot\mathbf{k}, \tag{4}$$

where $\mathbf{i}$, $\mathbf{k}$ are the unit vectors of the Cartesian coordinate system.

Then, after averaging in Eq. (3) over the surface S^* of the nanoparticles, we obtain the following form of the surface

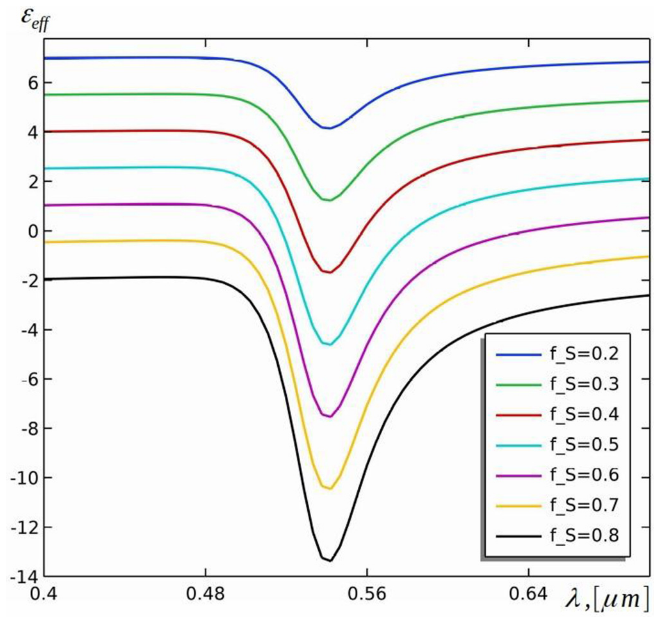

FIG. 2. Dependence of effective anchoring energy of NLC with the surface on the incident light wavelength λ. $f_S = 0.2$ (1), 0.3 (2), 0.4 (3), 0.5 (4), 0.6 (5), 0.7 (6), and 0.8 (7) (top to bottom).

free-energy density F_S:

$$F_S = -\frac{1}{2} W_{\text{eff}} \cos^2\theta, \tag{5}$$

where the density of effective surface free energy is

$$W_{\text{eff}} = (1 - f_s)W - \gamma f_s \frac{\varepsilon_a |E_0|^2}{4\pi}\left(\frac{\alpha^2}{5} + 1\right)d. \tag{6}$$

Figure 2 shows the dependence of the dimensionless effective anchoring energy $\varepsilon_{\text{eff}} = W_{\text{eff}}L/K_3$ of NLC with the substrate surface on the wavelength λ of the incident light, calculated for 5CB at different degrees of filling of the surface with gold nanoparticles. The calculations are performed for the cell thickness $L = 10\ \mu$m, anchoring energy $W = 10^{-5}$ J/m^2, amplification coefficient of the local electric field by the ensemble of gold nanoparticles $\gamma = 10$ and the intensity of the incident light wave $I = 100$ W/cm^2. Here and further, the dependence of the dielectric permeability of NLC [54] and gold nanoparticles [55] on the wavelength λ is taken into account. Based on the results of calculations, the decrease of ε_{eff} at a wavelength of $\lambda \approx 545$ nm of incident light corresponds to the region of plasmon resonance in the ensemble of gold nanoparticles in 5CB [48]. In this case, the local electric field near the surface with gold nanoparticles reaches the highest value, because the denominator $\varepsilon_1(\lambda) + 2\varepsilon_m(\lambda)$ of the coefficient α in the expression for $\mathbf{E}_{\text{loc}}$ (2) reaches the smallest value. It is expected that increasing the degree of coverage of the substrate with gold nanoparticles leads to a significant dropping down of the curve $\varepsilon_{\text{eff}}(\lambda)$ in the region of plasmon resonance.

To construct the free energy Φ (1) of the system, an explicit form of the vector $\mathbf{E}$ of the electric field in the bulk is required. As we have already mentioned above, in the calculations we neglect the impact of the local electric field of the ensemble of gold nanoparticles on the director in the bulk of the cell. We assume the light field has nonzero components E_x and E_z, and the component $E_y = 0$. Using the explicit expression (4) of the director, the free energy (1) of the cell can be written as

$$\Phi = \int_V \left[\frac{1}{2}(K_1 \sin^2\theta + K_3 \cos^2\theta)\theta_x'^2 - \frac{\varepsilon_a}{16\pi}(|E_z|^2 \sin^2\theta + |E_x|^2 \cos^2\theta + (E_x E_z^* + E_z E_x^*)\sin\theta\cos\theta)\right] dV - \frac{1}{2}\int_S W_{\text{eff}} \cos^2\theta \, dS. \tag{7}$$

The total free energy (7) minimization results in the following Euler-Lagrange equation:

$$(K_1 \sin^2\theta + K_3 \cos^2\theta)\theta_{xx}'' + (K_1 - K_3)\theta_x'^2 \sin\theta\cos\theta + \frac{\varepsilon_a}{16\pi}[(|E_z|^2 - |E_x|^2)\sin 2\theta + (E_x E_z^* + E_z E_x^*)\cos 2\theta] = 0$$

and boundary conditions to it

$$[(K_1 \sin^2\theta + K_3 \cos^2\theta)\theta_x' - W_{\text{eff}} \sin\theta\cos\theta]_{x=0} = 0, \quad \theta|_{x=L} = 0. \tag{8}$$

We are interested in the behavior of the system at the values of the electric field of the light wave close to the threshold of the director reorientation. To find the light intensity threshold value, we linearize Eq. (8) and the boundary conditions (8). The equation $\operatorname{div}\mathbf{D} = 0$ yields

$$k_x D_x = k_x(\varepsilon_{xx}E_x + \varepsilon_{xz}E_z) = 0. \tag{9}$$

Hence, with linear accuracy by θ we obtain

$$E_x = -\varepsilon_a \theta E_z/\varepsilon_\parallel. \tag{10}$$

Taking into account the relationship between the components E_x and E_z in the bulk, Eq. (8) in the linear by θ approximation transforms into

$$\theta_{xx}'' + \frac{\varepsilon_\perp}{\varepsilon_\parallel}\frac{\varepsilon_a E_0^2}{8\pi K_3}\theta = 0, \tag{11}$$

and corresponding boundary conditions (8) now read

$$[K_3\theta_x' - W_{\text{eff}}\theta]_{x=0} = 0, \quad \theta|_{x=L} = 0. \tag{12}$$

Requiring the solution of Eq. (11) to satisfy the boundary conditions (12), we obtain the equation to determine the threshold of the orientational instability:

$$\varkappa \cos\varkappa + \left[(1 - f_s)\varepsilon - \frac{2}{\pi}\left(\frac{\alpha^2}{5} + 1\right) f_s \frac{\varepsilon_\parallel}{\varepsilon_\perp}\frac{d}{L}\varkappa^2\right]\sin\varkappa = 0, \tag{13}$$

where $\varkappa = \frac{\pi E_0}{E_{0\text{th}}^\infty}$, $E_{0\text{th}}^\infty = \frac{\pi}{L}\sqrt{\frac{8\pi K_3}{\varepsilon_a}\frac{\varepsilon_\parallel}{\varepsilon_\perp}}$ is the Fredericks transition threshold with infinitely strong anchoring of NLC with the substrate surfaces in the absence of gold nanoparticles. Note that when $f_s = 0$ Eq. (13) transforms into the well known expression for threshold in the system without gold nanoparticles at the cell substrate [1].

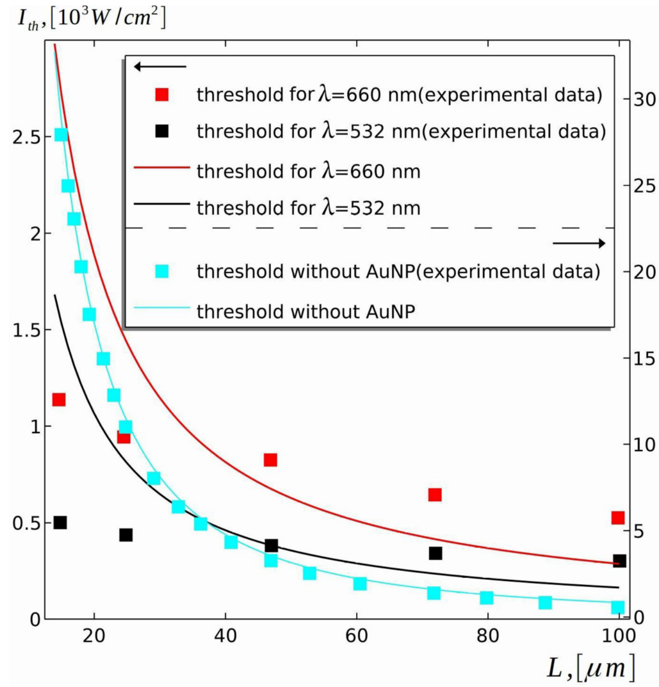

FIG. 3. Calculated and experimental dependencies of the OFT threshold intensity I_{th} on the cell thickness L.

Figure 3 demonstrates the comparison of the threshold intensity $I_{\text{th}} = c|E_{0\text{th}}|^2/(8\pi)$ found from Eq. (13) with experimental data from Ref. [48]. The calculations were performed for the parameters taken from Ref. [48] for the 5CB cell at the wavelengths of incident light $\lambda = 532$ and 660 nm [54]. The degree of filling of the substrate surface with gold

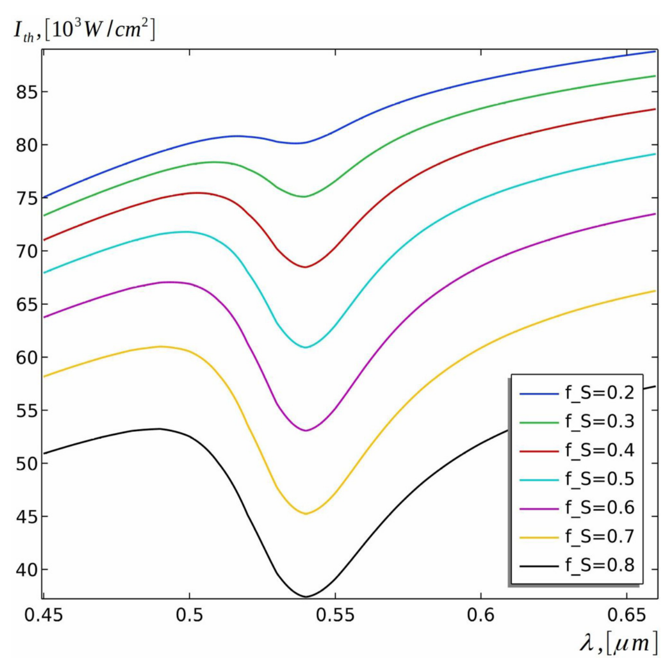

FIG. 4. Dependence of the threshold intensity I_{th} of OFT on the wavelength λ of incident light. $f_S = 0.2$ (1), 0.3 (2), 0.4 (3), 0.5 (4), 0.6 (5), 0.7 (6), and 0.8 (7) (top to bottom).

nanoparticles was $f_s = 0.5$. As one can see, with increasing cell thickness, the threshold light intensity decreases. However, this reduction is not as fast as in the absence of gold nanoparticles on the substrate surface. The accuracy of the model deteriorates as the cell thickness decreases, which may be a consequence of nonlinear optical phenomena [31,35–39] in the NLC media around gold nanoparticles when the frequency of an incident light wave is close to the plasmon resonance frequency. The impact of an optical nonlinearity becomes more noticeable at small cell thicknesses.

Figure 4 shows the calculated dependence of the threshold intensity I_{th} of OFT on the wavelength λ of incident light for different degrees f_s of covering the surface with gold nanoparticles. The calculations were performed for a 5CB cell of thickness $L = 10$ μm. Similarly to the dependence $\varepsilon_{\text{eff}}(\lambda)$ of the effective anchoring energy on the wavelength of incident light, the threshold of orientational instability I_{th} declines in the region of plasmon resonance. Thus, a significant increase in the local electric field in the ensemble of gold nanoparticles accelerates the development of the orientational instability of the director's field in the bulk of the NLC cell, which leads to lowering the threshold of the orientational instability threshold of the director's field. Increasing the degree of filling of the surface with gold nanoparticles leads to a decrease of the orientation instability threshold.

III. DISCUSSION

We developed the theoretical model of light-induced orientation instability of NLC director in a hometropically oriented cell with an ensemble of gold nanoparticles randomly deposited on the surface of one of the bounding substrates. The local electric field, significantly amplified by plasmon resonance in the ensemble of gold nanoparticles, reorients the NLC molecules of the near-surface layer. It reduces the effective anchoring energy of nematic with the substrate surface covered with gold nanoparticles. As a result, there is a significant decrease of the OFT threshold intensity. Therefore, the plasmon resonance that occurs in the ensemble of gold nanoparticles, opens the possibility for optical modulation of the effective anchoring energy of NLC with the substrate.

We propose a model of surface free-energy density of NLC, which describes the impact of the local electric field on the director of the near-surface layer of nematics near the surface with gold nanoparticles. Within the proposed model, the value of the threshold intensity of OFT was found and its dependence on the system parameters, in particular, the wavelength of incident light and the degree of filling of the surface with the nanoparticles, was investigated. As it follows from the comparison of theoretical results with experimental data [48], the proposed model describes well enough the decrease of the OFT threshold value due to the phenomenon of plasmon resonance in the ensemble of gold nanoparticles.

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