

Thermodiffusive photorefractive phenomena in liquid crystals

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We present a mechanism of photorefractivity that does not require photoconductivity for its existence. This mechanism works in light absorbing liquids that contain ions. The light-induced separation of charges takes place due to thermal diffusion. The effect is proportional to the power of absorbed light and can become stronger than conventional diffusive photorefractivity in liquid crystals. The thermodiffusive space charge field may acquire a component along the propagation direction of the light. It is shown that superimposition of thermal and photorefractive modulations of material parameters leads to the possibility of generation of orientational gratings that are spatially resonant with the wave's interference pattern. The results obtained may have large practical importance extending the class of photorefractive materials and offering possibilities of control of photorefractive optical phenomena. [S1063-651X(98)10710-9]

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I. INTRODUCTION

A breakthrough in the search for different photorefractive materials occurred with the prediction and discovery of photorefractive liquid crystals (LCs) [1–4]. Photorefractive LCs solve two key problems: First, the voltage necessary to apply to the photorefractive material for realization of wave mixing was reduced from kilovolts to volts and, second, the modulation of the refractive index of the material can be as large as 0.2. Both achievements are due to the unique electro-optical and mechanical properties of LCs [5–7].

The difference between the diffusion constants D^+ and D^- of photogenerated positive and negative ions is the reason of charge separation and space charge formation in photoconductive LCs [1,2]. In the case of the solution of the dye R6G in 4-pentyl-4'-cyanobiphenyl, $\nu = (D^+ - D^-)/(D^+ + D^-) \approx 0.02$. Getting larger ν is the main opportunity of further enhancing the photorefractive field formed due to photogeneration and subsequent diffusion of charges [8–10].

In the present article we will discuss a principally different mechanism of creating space charge fields that may be functional even in nonphotoconductive LCs. This thermodiffusive mechanism may become dominant for higher power laser beams used for recording of holographic gratings in photorefractive polymer dispersed LCs [11,12].

II. THERMAL DIFFUSION AS AN EFFECTIVE MECHANISM OF CHARGE SEPARATION

Strong spatial gradients of temperature can be obtained in laser beams acting on an absorbing medium. Therefore, the thermal diffusion (or Soret effect, as it is known for liquid

solutions [13,14]) can essentially modulate the concentration of ions present in a liquid. First, this process does not require photoinduced generation of charges and leads to a space charge modulation independent in the origin of ions in the liquid. Second, the thermal diffusion may become a stronger driving force than the mass diffusion for strong temperature gradients. Third, the difference in the thermal diffusion constants may be more substantial than the difference between the mass diffusion constants for the same ions.

To qualitatively characterize the thermal diffusion mechanism of space charge formation, let us write down the expression for the flux density \mathbf{J} of ions in the LC in the presence of temperature gradients:

$$\begin{aligned} \mathbf{J}^+ &= -D^+ \nabla n^+ - n^+ D_T^+ \nabla T + n^+ \mu^+ \mathbf{E}, \\ \mathbf{J}^- &= -D^- \nabla n^- - n^- D_T^- \nabla T - n^- \mu^- \mathbf{E}. \end{aligned} \quad (1)$$

In Eq. (1), T is the temperature, D is the coefficient of mass diffusion, D_T is the thermal diffusion coefficient, n is the density of ions, \mathbf{E} is the space charge field, and μ is the magnitude of the mobility of the ions. The superscripts indicate the sign of the ions. The positive value of the thermal diffusion constant D_T indicates that the particles are driven out from the region of high temperature towards the cooler regions.

In the steady state, the fluxes are absent and we arrive at the following system of equations to be solved:

$$\begin{aligned} -\nabla n^+ - n^+ S_T^+ \nabla T + \frac{\mu^+ n^+}{D^+} \mathbf{E} &= 0, \\ -\nabla n^- - n^- S_T^- \nabla T - \frac{\mu^- n^-}{D^-} \mathbf{E} &= 0, \end{aligned} \quad (2)$$

$$\text{div } \epsilon \mathbf{E} = 4\pi e(n^+ - n^-).$$

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Equation (2) allows us to express the space charge field through the modulation of the ion concentration and the temperature

$$\mathbf{E} = \frac{1}{n^+ \mu^+ + n^- \mu^-} [(D^+ \nabla n^+ - D^- \nabla n^-) + (D_T^+ \nabla n^+ - D_T^- \nabla n^-) \nabla T]. \quad (3)$$

As the first approximation, one can evaluate the space charge field from Eq. (3) by assuming $n^+ \approx n^- = n$ [1,2]. Making use also of the circumstance $D^+ \approx D^- = D$ and Einstein's relation $\mu = eD/k_B T$, Eq. (3) can be rewritten in the form

$$\mathbf{E} = \mathbf{E}_D + \mathbf{E}_{TD}, \quad (4)$$

where

$$\mathbf{E}_D = \nu \frac{k_B T}{2e} \frac{\nabla n}{n} \quad (5)$$

and

$$\mathbf{E}_{TD} = s \frac{k_B T}{2e} \nabla T. \quad (6)$$

The first term in Eq. (4), \mathbf{E}_D , yields in the conventional diffusive space charge field in the case of photogeneration of charges. The magnitude of \mathbf{E}_D [Eq. (5)] is thus determined by the difference in the diffusion constants of oppositely charged particles $\nu \approx (D^+ - D^-)/2D$. The second term in Eq. (4), \mathbf{E}_{TD} , is due to thermal diffusion and its magnitude [Eq. (6)] is proportional to the difference in the so-called Soret-Ludwig constants of oppositely charged particles $s = (D_T^+ - D_T^-)/D$.

The temperature variations induced by the interfering light beams can easily be determined from the temperature conductivity equation in the steady state:

$$\chi \nabla^2 T = - \frac{\sigma I}{\rho C_p},$$

where χ is the coefficient of the temperature conductivity, ρC_p is the specific heat capacitance of the LC per unit volume of the material at fixed pressure, and σ is the absorption constant.

Let us represent the spatial variation of the intensity of radiation I in the interfering light waves in the form

$$I = I_0(1 + m \cos \mathbf{q} \cdot \mathbf{r}), \quad (7)$$

where $I_0 = I_1 + I_2$ is the total average intensity of the interfering light beams, \mathbf{q} is the wave vector of the interference pattern, and $m = 2(I_1 I_2)^{1/2}/(I_1 + I_2)$ is the depth of modulation. The homogeneous part of the illumination will result in an average increase of the temperature, determined by the boundary conditions. Such a temperature increase may modify the material parameters; however, it will not yield in any redistribution of ions. The spatially modulated part of the temperature is equal to

$$\delta T = \frac{m \sigma I_0}{\chi \rho C_p q^2} \cos \mathbf{q} \cdot \mathbf{r} \quad (8)$$

if the effect of boundary conditions for the temperature is neglected. The magnitude of the space charge field generated due to the thermal diffusion can now be found to be equal to

$$E_{TD} = s \frac{k_B T}{2e} \frac{m \sigma I_0}{\chi \rho C_p q} \sin \mathbf{q} \cdot \mathbf{r}. \quad (9)$$

The characteristic feature of the thermodiffusive (TD) space charge field is its proportionality to the intensity of radiation and to the absorption constant and inverse proportionality to the wave vector of the interference grating.

III. EVALUATION OF THE SPACE CHARGE FIELD

To underline the characteristic features of the TD space charge field, let us compare it with the conventional diffusive photorefractive field E_D [2]:

$$\mathbf{E}_D = \nu \frac{k_B T}{4e} \frac{1}{I} \nabla I \approx - \nu m \frac{k_B T}{4e} \mathbf{q} \sin \mathbf{q} \cdot \mathbf{r}. \quad (10)$$

The independence of the diffusive space charge field on the concentration of photogenerated charges and hence on the intensity of radiation holds only for the case when the photoconductivity is much stronger than the dark conductivity. The direct proportionality of the field to the wave vector of the interference grating is a direct consequence of the assumption about local generation of charges.

While E_D saturates for the intensity of radiation needed to overcome the dark conductivity, the TD field can be effectively controlled with the aid of the incident intensity and the absorption constant. Comparing Eqs. (9) and (10), we find that the strength of the TD field becomes comparable to E_D for the rate of absorbed power per unit volume

$$\sigma I_0 \geq \frac{2 \pi^2 \nu \chi \rho C_p}{s \Lambda^2}, \quad (11)$$

where $\Lambda = 2\pi/q$ is the spatial scale of the interference pattern.

Let us carry out numerical evaluations for $\Lambda = 100 \mu\text{m}$ using the following typical material parameter values: $\nu \sim 10^{-2}$, $\chi \sim 10^{-3} \text{ cm}^2/\text{s}$, and $\rho C_p \sim 1 \text{ J/cm}^3\text{K}$. Assuming also that s is an order of magnitude smaller than an average value of the Soret constant $s \sim 10^{-3} \text{ K}^{-1}$ while $S_T \sim 10^{-2} \text{ K}^{-1}$ [15–17], we get $\sigma I_0 \approx 3 \times 10^3 \text{ W/cm}^3$. Such a value of the absorbed power density is readily achieved if a beam of intensity of about 30 W/cm^2 is interacting with a material of an absorption constant $\sigma = 100 \text{ cm}^{-1}$. By that, the overall change in the temperature of the medium turns to be reasonably small: $\delta T \sim \sigma I \tau / \rho C_p \sim 6 \text{ K}$, given $I = I_0(1 + m) \approx 2I_0$ and the typical thermal relaxation time $\tau \sim 1 \text{ ms}$.

IV. THE NORMAL COMPONENT OF THE SPACE CHARGE FIELD IN ABSORBING MEDIA

All the above discussion assumed modulation of the light intensity in two dimensions only, in the plane of the LC cell.

This is obviously not correct in the presence of strong absorption. With $\sigma=10^2 \text{ cm}^{-1}$, the intensity of the light attenuates remarkably (2.7 times) in a distance of $100 \mu\text{m}$, the typical thickness of the LC cells. This attenuation, however, is not merely a quantitative modification of the estimations made above, but leads to important possibilities. Namely, thermal diffusion may separate the charges along the thickness of the cell yielding in a space charge field along the normal to the LC layer.

To describe and evaluate this effect, let us represent the intensity of the light beam in the medium as

$$I(z)=I_0\exp(-\sigma z), \quad (12)$$

where z is the axis of the Cartesian coordinate system that is perpendicular to the cell plates and coincides with the direction of propagation of the light beam. Replacing q with σ and assuming $m=1$ in Eq. (10), for the normal component of the diffusive space charge field we can easily get

$$E_{zD}=-\nu \frac{k_B T}{e} \sigma. \quad (13)$$

To determine the normal component of the TD space charge field, let us solve the temperature conductivity equation for the one-dimensional problem and with account of the expression for the light intensity (12). Imposing boundary conditions $T(z=0)=T(z=L)=0$, we get

$$\frac{\partial T}{\partial z} = \frac{I_0}{\chi \rho C_p} \left[e^{-\sigma z} - \frac{1}{\sigma L} (1 - e^{-\sigma L}) \right] \approx \frac{I_0}{\chi \rho C_p} \left(e^{-\sigma z} - \frac{1}{\sigma L} \right). \quad (14)$$

Consequently,

$$E_{z \text{ TD}} = s \frac{k_B T}{2e} \frac{I_0}{\chi \rho C_p} \left(e^{-\sigma z} - \frac{1}{\sigma L} \right). \quad (15)$$

Comparing the normal components of the diffusive and TD space charge fields, we find that the latter becomes stronger if the absorbed power density is larger than

$$\sigma I_0 \geq \nu \chi \rho C_p \sigma^2 s^{-1}. \quad (16)$$

For typical parameter values accepted above, $\sigma I_0 \geq 10 \text{ W/cm}^2$.

V. REORIENTATION OF LCs AND RESONANT GRATINGS DUE TO MODULATION OF THE ORDER PARAMETER

The magnitude of reorientation of a nematic LC (NLC) is determined by variational equations for the components of the NLC director. Let us describe the orientation of the director with a single angle θ such as $\mathbf{d}=(\sin \theta, 0, \cos \theta)$. Then

$$\frac{\partial^2 \theta}{\partial x^2} + \frac{\partial^2 \theta}{\partial z^2} = -\frac{\Delta \varepsilon}{4 \pi K} [(E_x^2 - E_z^2) \sin \theta \cos \theta + E_x E_z \cos 2 \theta], \quad (17)$$

where E_x, E_z are the components of the electric field, $\Delta \varepsilon$ is the dielectric constant of the NLC at the frequency of the acting electric field, and K is the elastic constant. Below we will consider

$$E_z=0, \quad E_x=E_{x0} \sin q x$$

to reveal the possibilities implied in the superimposition of thermal and photorefractive gratings.

Even small reorientation of the NLC yields a large change in the refractive index due to its large optical anisotropy. Therefore, we will be interested in solving Eq. (17) linearized over θ . Let the NLC has a pretilt $\theta=\theta_0$ at the undistorted state. Then the linearized equation for small perturbations $\theta=\theta_0+\delta\theta$ will obtain the form

$$\frac{\partial^2 \theta}{\partial x^2} + \frac{\partial^2 \theta}{\partial z^2} = -\frac{\Delta \varepsilon \sin \theta_0 \cos \theta_0}{4 \pi K} E_{x0}^2 \sin^2 q x, \quad (18)$$

Let us assume strong anchoring at the boundaries: $\delta\theta(z=0)=\delta\theta(z=L)=0$. The temperature dependence of the material parameters of NLC may allow recording of a reorientation grating of a wave vector q also in the case when the quadratic combination of the electric fields that give rise to the reorientation of the director has a spatial frequency $2q$. Namely, the interfering light beams in an absorbing medium give rise not only to the space charge modulation, but also to the modulation of the material parameters of the LC. The relevant parameter in our problem is $\Delta \varepsilon/K$. Since $\Delta \varepsilon \sim Q$ and $K \sim Q^2$, where Q is the order parameter of the material, we get that the ratio $\Delta \varepsilon/K \sim Q^{-1}$ and may strongly increase near the transition point of the LC to the isotropic phase.

Since the temperature profile follows the profile of the interference pattern, let us take

$$\eta = \frac{\Delta \varepsilon}{K} = \eta_0 + \eta_m \cos q x \quad (19)$$

in the linearized Eq. (18). Thus the presence of a thermal modulation of the material parameters allows the photorefractive field at the frequency q to create a torque acting on the NLC orientation at the spatial frequency q . The equation that determines the amplitude of the grating at the wave vector q has the form

$$\frac{\partial^2 \theta_q}{\partial x^2} + \frac{\partial^2 \theta_q}{\partial z^2} = -\eta_m \sin \theta_0 \cos \theta_0 \frac{E_{x0}^2}{16 \pi} \sin q x. \quad (20)$$

The solution of Eq. (20) is

$$\theta_q = \eta_m \sin \theta_0 \cos \theta_0 \frac{E_{x0}^2}{4 \pi q^2} \times \left[1 - e^{-qz} - \frac{\sinh qz}{\sinh qL} (1 - e^{-qL}) \right] \sin q x. \quad (21)$$

It is important that η_m not only can be very large close to phase transition points (nematic-isotropic and nematic-smectic), but also can reveal a more intricate temperature dependence, yielding a shift between the gratings of the waves' interference pattern and the orientation.

VI. PHOTOREFRACTIVE STORAGE IN LC COMPOSITES

Though the phenomena under discussion can take place in any absorbing liquid, use of LCs has important advantages. Apart from the above-mentioned possibility of large refractive index modulation with only several volts of applied voltage, LCs provide unique possibilities of material architecture both for the use of optical phenomena and with the aid of optical phenomena. Thus polymer dispersed liquid crystals (PDLCs) were designed as materials with effectively controllable light scattering properties [18]. The interference pattern of light was used to create controllable gratings of PDLCs [19,20].

Such composite materials can effectively be used to achieve different photorefractive applications [11,12]. The polymer network or the polymer matrix can play the role of traps for charges and realize a storage effect. In the recording process, the polymer matrix is “melted” and the charge specific diffusion separates the charges creating the space charge field. At a high temperature, however, the LC is in its isotropic state and the space charge field will have a negligible effect on the refractive index since the typical strength of the electric field needed to influence the order parameter of the isotropic LC is rather high. In the case of molecules carrying a permanent dipole, it can be estimated as $E \sim k_B T / ea$, where a is the size of the molecule. Therefore, $E \sim 10^6$ V/cm.

Thus, in the recording process, the laser beam yields the formation of the space charge field, but there is no considerable modulation of the refractive index while the beam is on and the temperature of the system is high. With switching off the laser beam, two processes start to take place simultaneously: phase separation of the mixture into the PDLC and relaxation of the space charge field due to the diffusion of ions. The cooling down of the system takes place during $\tau_T \sim L^2 / \pi^2 \chi \sim 10^{-2}$ s. The smearing out of the temperature grating is a process about two orders of magnitudes faster due to the smallness of the grating spacing Λ compared to the cell thickness (or the size of the beam).

The diffusion time of the ions shall be evaluated more accurately, taking into account the existence of the space charge field that influences the motion of the ions. Let us proceed from the continuity equation for the charges of different signs:

$$\frac{\partial n^+}{\partial t} = -\nabla \cdot \mathbf{J}^+, \quad \frac{\partial n^-}{\partial t} = -\nabla \cdot \mathbf{J}^-, \quad (22)$$

where the fluxes have been defined in Eq. (1). Let us deal with the difference in the charge concentration $n^+ - n^-$:

$$\frac{\partial(n^+ - n^-)}{\partial t} = -\nabla \cdot (\mathbf{J}^+ - \mathbf{J}^-). \quad (23)$$

To write down the difference in the flux density, let us take into account the following circumstances. First, one can neglect with the difference in diffusion coefficients for charges of different signs. This difference was essential to get charge separation in a photoconductive LC; however, it is only a minor correction when the estimation of the relaxation times are concerned. As mentioned above, the temperature can be considered to be homogeneous due to fast wash-

ing out of its spatial modulation. Assuming also $n^+ + n^- \approx 2n_0 \gg n^+ - n^-$, we will eventually get

$$\mathbf{J}^+ - \mathbf{J}^- = -D\nabla(n^+ - n^-) + 2\mu n_0 \mathbf{E}. \quad (24)$$

Equation (24), with account of the equation of the electrostatics (2), is transformed now into

$$\frac{\partial E}{\partial t} = D\nabla^2 E - \frac{8\pi e^2 n_0 D}{\epsilon k_B T} E. \quad (25)$$

Thus the space charge field with the wave vector q relaxes as

$$\frac{\partial E_q}{\partial t} = -\left(\frac{1}{\tau_D} + \frac{1}{\tau_E}\right) E_q, \quad (26)$$

where

$$\tau_D = \frac{1}{Dq^2}, \quad \tau_E = \frac{\epsilon k_B T}{8\pi e^2 n_0 D} \approx \frac{1}{3\pi n_0 D R_S}, \quad (27)$$

with R_S having the physical meaning of the distance where the binary interaction potential energy of ions is balanced by their kinetic energy of Brownian motion. The characteristic diffusion time τ_D describes the relaxation of the concentration perturbations for neutral molecules only. For perturbations of comparable spatial scale, τ_D is much larger than the temperature relaxation time τ_T , $\tau_D / \tau_T = \chi / D \sim 10^3$. Thus τ_D is still an order of magnitude smaller than τ_T even for perturbations with the spatial scale 10 times smaller than the thickness of the LC cell.

For charged molecules, however, the attraction of charges of opposite signs can become the dominant mechanism of relaxation of the space charge field. The electrostatic relaxation time of the space charge field τ_E is larger than or comparable to the thermal relaxation time up to concentration of charges

$$n_0 < \frac{\chi}{D R_S L^2}. \quad (28)$$

For typical parameter values $\epsilon = 10$, $D = 10^{-6}$ cm²/s, and $L = 10^{-2}$ cm, we get $n_0 \sim 10^{13}$ cm⁻³. Such a concentration of charges is still large enough to cause strong photorefractive effects [1,2].

Thus experimental situations where the space charge field is relaxing slower than the temperature of the material can straightforwardly be realized. In such a situation, the LC will cool down to its mesophase and the mixture will separate into LC microdroplets dispersed in a polymer in conditions of the action of a spatially modulated electric field. It is well known that such a process may lead to orientation of the LC in the polymer matrix [21,22]. Moreover, such an orientation has a memory effect: It is recovered when heating the system to the isotropic phase with consequent cooling down to the mesophase.

VII. SUMMARY

We have revealed a mechanism of formation of space charge fields: thermal diffusion. This mechanism works in nonphotoconductive materials and can be dominant even in

photoconductive systems. We showed that absorption can lead to the creation of a space charge field along the direction of light propagation and this field can be as strong as the conventional space charge field in the particular material.

Thermal diffusion may lead to interesting phenomena of recording of amplitude and phase gratings in electrically neutral solutions, with no ions or photogenerated charges at all. Actually, processes of amplitude grating formation due to thermal diffusion in systems containing absorbing centers have been previously reported [23], particularly, for magnetic fluids [24]. As suggested in [25], the thermal diffusion may be the reason for the earlier observations of aberrational self-focusing effects in magnetic fluids [26]. The component of the thermal diffusion along the normal to the boundaries of the LC cells and/or the related normal component of the space charge field may result in the deposition of particles and molecules on the substrate, thus modifying the boundary conditions and resulting in a memory effect [4,27].

For dichroic dyes dissolved in NLCs, the light beam strongly influences the orientation of the dye and its interac-

tion with the host LC. This effect is widely explored nowadays in obtaining amplification of the orientational optical nonlinearity of LCs; see [28,29]. Thus we shall expect that the redistribution of the dichroic dye concentration is accompanied by the redistribution of the molecular axes as well. Redistribution of the dye concentration will evidently modulate also the orienting action of the dye molecules on the LC molecules giving rise to a reorientation pattern that can be revealed as an anisotropic grating.

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