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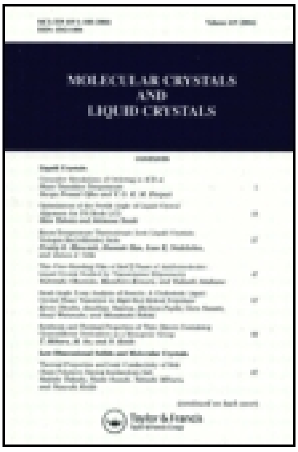

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Thermal Optical Nonlinearity of Suspension of Absorbing Particles in Liquid Crystal

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We studied the thermal optical nonlinearity in a suspension of ultra-fine absorbing particles in a nematic liquid crystal. Each particle "works" as a light-controlled "heater" of surrounding LC that results in change of the refractive index of the suspension. If the distance between absorbing particles in the suspension is less than the characteristic length of thermodiffusion, the thermal and refractive index profiles are close to those of a homogeneously absorbing medium. Therefore, thermal optical nonlinearity in suspensions of absorbing ultra-fine particles in LC can produce the same result as that in dye-doped LCs. The advantage of the proposed system is the absence of the effects, which usually accompany light-induced heating.

Keywords: liquid crystal; inorganic particles; suspension; optical thermal nonlinearity.

INTRODUCTION

Thermal nonlinearity of dye-doped liquid crystals (LC) is characterised with the anomalously high parameter of cubic nonlinearity $\varepsilon_2 \sim 10^{-5}$ esu and the fast optical response (10^{-3} - 10^{-4}s)[1]. It makes these systems very promising for different applications. At the same time, thermal

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nonlinearity usually is accompanied by other nonlinear mechanisms of refractive index change (orientational[2,3], conformational[4,5], photoorientational[6] nonlinearities, hydrodynamic instability, etc). This prevents wide application of thermal nonlinearity in real devices and makes it difficult to interpret the experimental results of thermal nonlinearity studies. Furthermore, these materials usually are not stable because of photo-degradation, especially at high light intensity. Therefore, the development of an effective and stable media for thermal nonlinearity applications is important. To get these difficulties over we propose to use suspensions of non-organic ultra-fine absorbing particles in liquid crystals. Each particle "works" as a light-controlled "heater" of the surrounding LC. If the distance between particles is less than the characteristic length of thermodiffusion, the thermal and refractive index profiles are close to those of a homogeneously absorbing medium. In this work we present a theory of the optical thermal nonlinearity in LC suspensions and report on the first observation of the effect of thermal nonlinearity in the suspension of V_2O_5 particles in a LC pentyl-cianobiphenyl (5CB).

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THEORY

In order to make a theoretical description of a LC with absorbing particles we consider a LC layer of the thickness L sandwiched between two surfaces and placed in the thermostat. Let us presume that LC does not absorb light, the size of the light absorbing particles is negligibly small, and their positions are distributed randomly. The cell is irradiated with a gaussian laser beam

$$\tilde{I} = \tilde{I}_0 e^{-r^2/R^2} ,$$

where R is the half width of gaussian, $r = \sqrt{x^2 + y^2}$ is the spatial co-ordinate, $\tilde{I}_0$ is the intensity of the light.

Our prime interests are in the distribution of the temperature in the bulk of LC, the refractive index modulation and, as a consequence, the far field light intensity distribution. Thus, the problem is divided into two parts: first, we consider thermal properties of LC and second we consider the propagation of a light beam through the LC layer.

We can model our problem by the equation of heat-conductivity, which in Cartesian coordinates is given by:

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$$a^2 \Delta T = -\widetilde{I}_0 e^{\frac{-x^2-y^2}{R^2}} \sum_{i=1}^{N} \delta(x-x_i)\delta(y-y_i)\delta(z-z_i),$$

where T is the temperature, $\Delta = \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right)$ is the laplacian, a is the coefficient of heat conductivity, N is the number of the particles and x_i, y_i, z_i are the coordinates of the particle with number *i*, chosen from the volume of the side *2R* and thickness *L*.

There are uniform boundary conditions for this equation:

$$T(z=0) = T(z=L) = 0,$$

which imply that the temperature of the surfaces is a constant.

The solution of this equation reads:

$$T(x,y,z) = \sum_{i=1}^{N} \sum_{k=1}^{\infty} \exp(-r_i^2 / R^2) \sin(\sqrt{\lambda_k} z) \sin(\sqrt{\lambda_k} z_i) * \\ * \frac{\widetilde{I}_0 L}{\pi a^2 k} K_0 (\sqrt{\lambda_k ((x-x_i)^2 + (y-y_i)^2)}) \quad (1)$$

where $\lambda_k = \left(\frac{k\pi}{L} \right)^2$, K_o is the modified Bessel function of the second kind.

As one can see from the FIGURES 1,2,3, in the case of the small number of the particles there is no symmetry in the temperature distribution and the temperature function cannot be described by a Gauss function. When the number of the particles increases, the temperature distribution becomes more symmetric and similar to a Gauss function.

According to the scalar theory of diffraction and the Huygens-Fresnel principle, the amplitude of the light field of a gaussian beam after the LC cell can be evaluated from the following integral:

$$U(x_f, y_f) = \frac{e^{ikz}}{i\lambda z} \iint_{\Sigma} U(x,y) \exp\left\{ i \frac{k}{2z} \left[x^2 + y^2 - 2(xx_f + yy_f) \right] \right\} dxdy \quad (2)$$

where x, y, x_f, y_f are the cartesian coordinates in the LC plane and in the screen respectively, z is the distance from LC cell to the screen, $k = \frac{2\pi}{\lambda}$, and:

$$U(x,y) = t_l(x,y)U_l(x,y) \tag{3}$$

is the local amplitude of the field just after LC.

$$U_l(x,y) \sim \exp\left(-r^2 / R^2\right) \tag{4}$$

is the local amplitude of the field just before LC.

$$t_l(x,y) = \exp(ik\Delta\phi(x,y)) \tag{5}$$

is the coefficient of transmission, where $\Delta\phi(r,\varphi)$ is the phase retardation at the out plane of the cell.

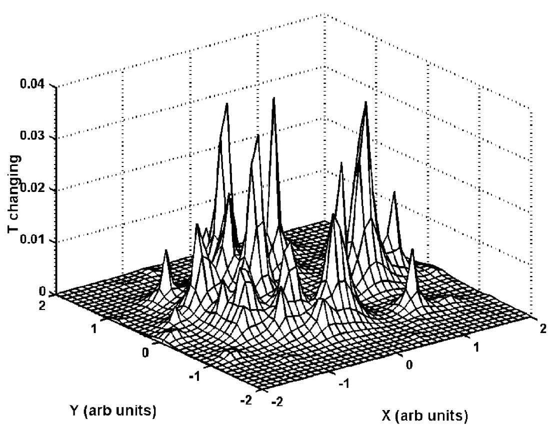

FIGURE 1. Spatial temperature distribution in the middle layer of the cell, *N=25*.

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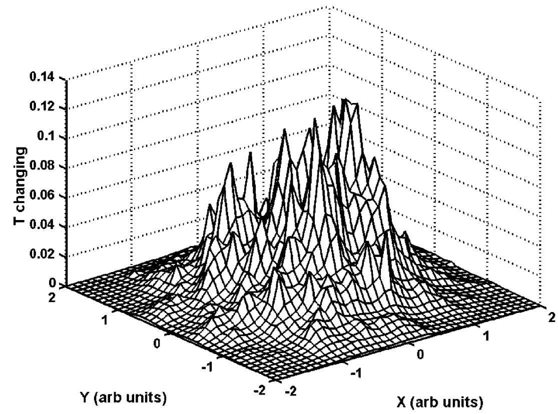

FIGURE 2. Spatial temperature distribution in the middle layer of the cell, N=*100*.

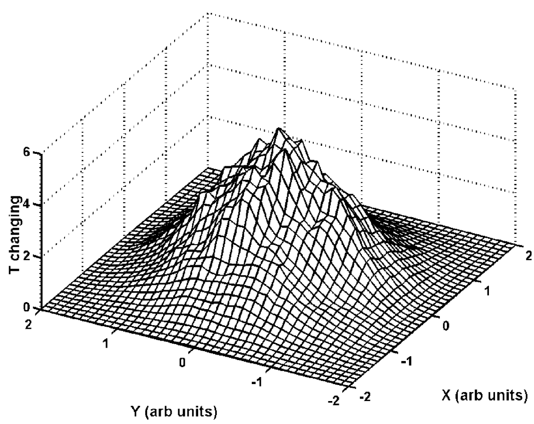

FIGURE 3. Spatial temperature distribution in the middle layer of the cell, N=250.

Change of the refractive index of LC caused by light-induced heating with a gaussian beam results in a spatial distribution of the phase retardation at the out plane of the cell:

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$$\Delta\phi(x,y) = \int_0^L dz\,\delta n(x,y,z) \tag{6}$$

The changes in the refractive index δn caused by the heated particles can be expressed as:

$$\delta n = \varepsilon_2^T \delta T / 2n$$

where ε_2^T is the parameter, which characterises thermal optical nonlinearity and associates with a change of the dielectric polarisability of a media with the light-induced heating:

$$n^2 = \varepsilon = \varepsilon_o + \varepsilon_2^T T(\tilde{I})$$

After the substitution (3), (4), (5), (6) into (2) one can evaluate the amplitude of the field:

$$U(x_f, y_f) \sim \iint_\Sigma \exp\left(-\frac{r^2}{R^2}\right) \exp\left(ik \int_0^L dz\, \delta n(x,y,z)\right) * \\ * \exp\left\{i\frac{k}{2z}\left[r^2 - 2(xx_f + yy_f)\right]\right\} dxdy \tag{7}$$

The integral (7) should be calculated numerically in order to find the distribution of the intensity.

The results of the numeric calculation of the far field intensity distribution for different intensities and different numbers of the particles *N* are presented in the Table 1.

The cells with the small number of the particles (*N=25*) do not show gaussian lens behaviour even at the high light intensity. At the low beam intensity these cells are transparent for the laser beam. As one can see, increasing the intensity changes the far field intensity distribution slightly in the cells with few particles. In this case the average distance between particles equals to *22* μm, which is much bigger than the length

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of thermodiffusion $l_T = 10\ \mu m$*. Irradiation of the cell with the moderate number of particles (*N=100*) results in an appearance of speckle-like structure due to micro-spatial irregularities of the refractive index nearby the particles. This number of the particles corresponds to the average distance of *10,5* μm and can be compared with the thermodiffusion length l_T. The cell with the large number of the particles (*N=250*) shows optical properties similar to the properties of nonlinear gaussian lens. Now the average distance for this number is *6,5* μm, which is smaller than the thermodiffusion length l_T. Due to the overlapping of the heated areas at the large particle concentration the micro-scale irregularities of the light intensity are weak in this case, and a characteristic aberration pattern of concentric rings appears. The number of the rings is multiply to $\Delta\phi = m\pi/2$ of the phase retardation at the center of the beam and increases with the increasing of the intensity [1,2].

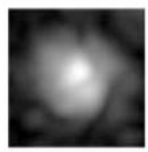
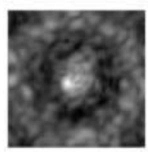
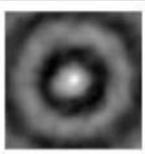
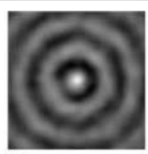
	$\tilde{I}_0 = 7\,mW$	$\tilde{I}_0 = 15\,mW$
N=25		
N=100		
N=250		

TABLE 1. Far field intensity distribution for different intensities and different number of the particles.

* The thermodiffusion length l_T was determined as the distance at which the temperature around the particle decreased in ten times. The value l_T was evaluated from expression (1).

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EXPERIMENT

The suspensions of V_2O_5 in a LC 5CB from EM Industry were prepared with different weight concentrations of vanadium oxide ($C = 0,2\text{-}5\%$). We found that the suspensions with the concentration $C > 2\%$ were not stable and the particles coagulated to large agglomerates of different shapes with the diameter 30 - 50 μm. Therefore, we carried out the experiments with the stable suspensions $C \leq 2\%$, in which the particles were homogeneously dispersed in the LC bulk. An average size of the particles in these suspensions was 3-5 μm. The average distance between particles in the suspension with $C = 2\%$ was 10 μm.

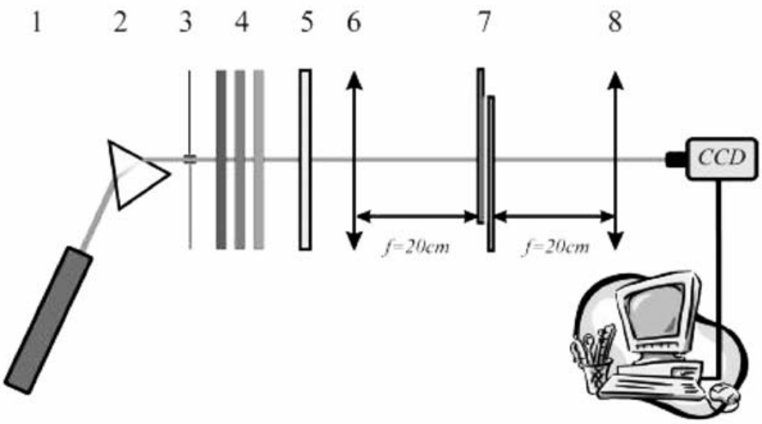


FIGURE 4. Experimental setup. 1 – Ar^+ laser; 2 – prism; 3 – diaphragm; 4 – polariser; 5 – filters; 6, 8 – lens; 7 – cell.

Besides of the thermal nonlinearity in the suspensions, the nonlinearity in pure 5CB and in the solution of the photo-stable dye pinavirdol in 5CB (1% by weight) were studied. The absorption of the dye-doped LC cell and the absorption of the cell with the 2% - suspension were (37 ± 1)%. The materials were sandwiched between two transparent glass substrates. Their inner surfaces were coated with rubbed layers of polyimide to provide a uniaxial director orientation in the whole cell. The thicknesses of the cells were in the range 51 – 55 μm.

The experimental scheme is depicted in FIGURE 4. The cells were irradiated by linearly polarised light beam from an Ar^+ laser ($\lambda = 488$ *nm*). The filters changed the intensity of light in the plane of the cells from 2,5 to 75 W/cm^2. The incident beam polarisation was adjusted parallel to the director orientation to avoid orientational nonlinearity. The far-field pattern was formed by the lens system and was detected by a CCD-camera connected to a computer.

The far field light-intensity distributions of the laser beam passing through the cells are presented in the TABLE 2.

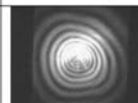
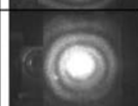
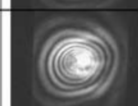
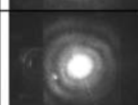
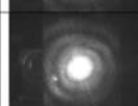
	Pure 5CB	5CB+0,2%V_2O_5	5CB+2%V_2O_5	5CB+1%Dye
75 W/cm^2				
60 W/cm^2				
50 W/cm^2				
40 W/cm^2				
30 W/cm^2				
25 W/cm^2				

TABLE 2. Far-field pattern for: 1) pure 5CB; 2) 5CB + 0,2% V_2O_5; 3) 5CB + 2% V_2O_5; 4) 5CB + 1% of dye.

As would be expected, no thermal non-linear effects were observed in the cells filled with pure 5CB, as both the LC and the substrates are transparent at the wavelength of the laser beam. In the cells filled with the suspension 5CB+0,2% V_2O_5 , which had the weak absorption (< 5%)

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we observed a strong light-induced speckle-like scattering around the beam ($\widetilde{I} > 40\ W/cm^2$).

In the cell with 2%-suspension even the low-intensive light ($\widetilde{I} \approx 5\ W/cm^2$) caused increasing of the beam divergence, which linearly depended on the light intensity (FIGURE 5, curve 3). Almost the same behaviour of the beam was observed in the cell with the light absorbing dye-doped LC (FIGURE 5, curve 2). At the intensities more than 30 W/cm^2, the ring-pattern characteristics to the formation of gaussian lenses were observed both in the 2%-suspension and dye-doped LC. The number of the rings linearly increased with the light intensity in both cases in accordance to the References [1,3].

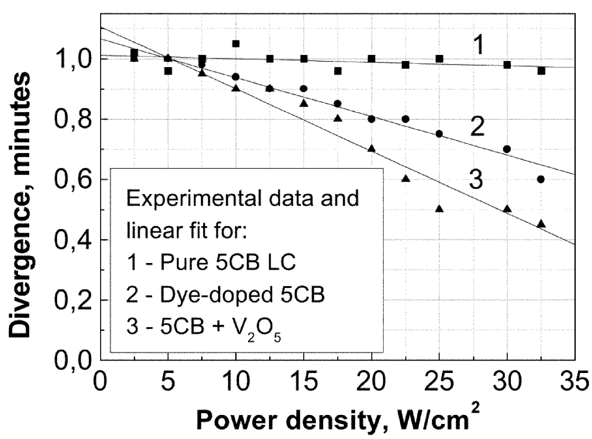

FIGURE 5. Light-induced changes of the beam.

The described experimental data agree with our theoretical predictions. At the low concentration of the absorbing particles, the light-induced heating is weak, temperature field is spatially inhomogeneous and far from a gaussian shape. Therefore, the light-induced heating results just in a local increasing of the temperature nearby of the particles. At the high light intensities it causes producing of chaotically distributed micro-lenses in the LC and the appearance of the light-induced speckle-like scattering. At the high concentration of the particles the heated areas around the particles overlap and the gaussian beam produces a gaussian-like temperature distribution in the LC bulk. As a result, a nonlinear gaussian lens is formed in the cell.

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CONCLUSIONS

We have shown that if the distance between absorbing particles in LC suspension is less than the characteristic length of thermodiffusion, the thermal and refractive index profiles are close to those of a homogeneously absorbing medium. Therefore, thermal optical nonlinearity in suspensions of absorbing ultra-fine particles in LC can produce the same result as that in dye-doped LCs. In the case of the suspension of 3-5 mμ particles of V_2O_5 in LC 5CB the characteristics of thermal nonlinearity become the same as for dye-doped LC at the concentration of the particles about 2%. Because the low concentration the particle does not increase the light scattering in the sample, the studied system looks like a good candidate for an efficient thermal-nonlinear medium. The advantage of the proposed system is the absence of the effects, which usually accompany light-induced heating.

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