

# Enhanced two-beam coupling in colloids of ferroelectric nanoparticles in liquid crystals

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We report on the first, to the best of our knowledge, studies of photorefractive in nematic liquid crystal (LC) doped with nanoferroelectric particles. We found the strong enhancement of two-beam coupling in the colloid of ferroelectric nanoparticles in LC. The effect originated from an increased birefringence of the colloid and a stronger LC reorientation torque. Our measurements allowed us to suggest that increased birefringence is caused by the contribution of polarizability anisotropy of the ferroelectric particles. Stronger reorientation torque is caused by the permanent dipole moment of the particles contributing to the dielectric anisotropy of the colloid  $\epsilon_a^{\text{col}}$ . The enhancement of two-beam coupling in LCs by doping with ferroelectric nanoparticles at extremely small concentration shows the strong potential of ferroelectric nanoparticles for improving the optical response of LCs, especially for those materials where a method of chemical synthesis has reached its limit.

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## 1. INTRODUCTION

Photorefractive in liquid crystals (LCs) has attracted significant attention since it is characterized by strong optical nonlinearities at low light intensities and voltage. The photorefractive effect in a LC could be initiated by light-induced charges in its volume [1–9], in the aligning layer [10–12], or at the interface between the LC and the aligning layer [13–19]. In volume-mediated photorefractive, the optical grating generates photocharges in the LC bulk that migrate within the LC to set up a dc space charge field. This field creates a torque on the LC director, forcing its reorientation and refractive index changes. In surface-mediated photorefractive, an interference pattern modulates charges on or in the aligning, photosensitive layer that leads to surface-induced torque in the LC bulk and results in a coupling between the recording light beams.

In this paper, we show that the efficiency of two-beam coupling in a nematic LC cell increases by doping LC with submicrometer ferroelectric particles. The idea of application of ferroelectric nematic colloids for optical processing arose after publications [20–27], in which the unique electro-optical and dielectric properties of these systems were observed. Doping LC with submicrometer particles at low concentrations (<1%) resulted in the strong increase of the dielectric and optical anisotropy and decrease in the Freedericksz transition voltage. Besides, a linear electro-optical response of the suspension suggested a break of the inversion symmetry in the colloid. All these factors allowed us to expect an enhancement of the orientation torque, which is responsible for the photorefractive effect in nematic LC.

## 2. EXPERIMENTS AND DISCUSSION

We used the combined LC cells made from two glass substrates covered with indium tin oxide electrodes. The thickness of the cell,  $L$ , was 16  $\mu\text{m}$ . A layer of polyvinylcarbazole (PVK) doped with  $\text{C}_{60}$  (weight concentration,  $c \approx 15\%$ ) was deposited on the substrates. This polymer was chosen as it provides a very efficient photorefractive effect [19]. The other substrate was covered by a polyimide layer. To achieve a homogeneous planar alignment of LC, both polymer layers were unidirectionally rubbed with velvet. Easy orientation axis on the PVK layer was orthogonal to the rubbing direction, and the easy axis on the polyimide layer was parallel to the rubbing. Therefore, to get homogeneous alignment, the directions of rubbing were mutually perpendicular on the substrates. The cells were then filled with either the LC or with the colloid of ferroelectric particles in the same LC at room temperature.

The nematic LC chosen was LC 18523 from Merck. This material was specially synthesized to have its refractive indices matched to silica with the aim of integration into fiber devices ( $n_e=1.51$  and  $n_o=1.46$  at  $\lambda=589$  nm). Hence, it is compatible and suitable for integration with fibers and optoelectronic devices. Indeed, this LC has been used in silica-on-silicon tunable Bragg gratings sensors and filters [28]. However, it has low birefringence, which limits its use in most applications that rely on the reorientation or modulation of LC orientation contrast. So far, no other low-refractive-index LCs with higher birefringence were successfully synthesized. Hence, our interest is pursuing nonsynthetic routes, namely, ferroelectric

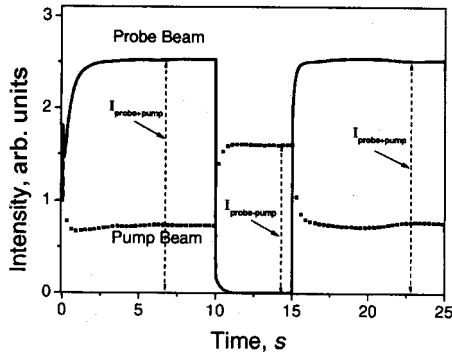


Fig. 1. Intensity exchange between the beams of equal intensities in the cell with the colloid.  $t=0$ ; 15 s, both beams are switched on.  $t=10$  s, pump beam is switched off.

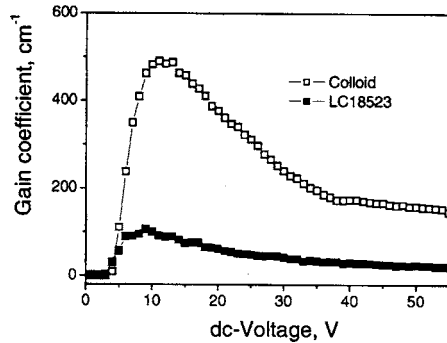


Fig. 2. Dependence of the gain coefficient on the applied voltage measured for the LC and the colloid.

nanoparticles colloid in this LC, to achieve an improved optical response.

We prepared the colloid by milling ferroelectric particles of  $\text{Sn}_2\text{P}_2\text{S}_6$  ( $\cong 1 \mu\text{m}$  size) mixed with oleic acid in a weight ratio of 1:2 according to the procedure described in the [20]. The characteristic size of the particles in the colloid was  $\sim 100$  nm, and their volume fraction,  $c_{\text{part}}$ , was  $\sim 0.3\%$  in the LC matrix.

The recording of the optical grating in LC cells was performed in a standard two-beam scheme [19]. Intensity gratings with a period  $\Lambda \approx 5\text{--}25 \mu\text{m}$ , was recorded in an LC cell via interference of two  $p$ -polarized beams at 543 nm from a He–Ne laser. The intensity of the incident beams in the plane of their intersection was  $I_0 = 17 \text{ mW/cm}^2$ . The data acquisition system allowed us to simultaneously monitor the intensity of transmitted as well as diffracted beams. A set of electromagnetic relays controlled both the application of an electric field to the cell's electrodes as well as shutters that were used to switch on and off the incident beams. A dc power supply and a waveform generator were used to apply either dc and ac electric fields.

We concentrated on the measurement of self-diffraction and two-beam coupling gain. The irradiation of the cells at the simultaneous application of the dc voltage resulted in the recording of a dynamic Raman–Nath diffraction grating and the appearance of the self-diffraction of the recorded beams. The characteristics of the self-diffraction and the asymmetric energy exchange between the recording beams for the pure LC and ferroelectric colloid were

qualitatively the same as the characteristics described earlier in [19]. The most efficient self-diffraction and beam coupling were observed when the cell was rotated away, so a normal of its incident surface did not coincide with the bisector of the angle between the recording beams. The optimum range of rotation angles was between  $20^\circ\text{--}30^\circ$ , where the diffraction efficiency exceeded 20%.

The recording of the grating was accompanied by the intensity exchange between the recording beams of the equal intensities. It was observed as the amplification of one of the recording beams at the expense of the depleting of the other one. The dynamic of the beam coupling in the colloid is depicted in Fig. 1 for  $\Lambda \approx 11 \mu\text{m}$ . To characterize the beam coupling, we measured the ratio  $G = I_{\text{probe+pump}}/I_{\text{probe-pump}}$ , where  $I_{\text{probe+pump}}$  is the intensity of the probe beam in the presence of the pump beam and  $I_{\text{probe-pump}}$  is the intensity of the probe beam in the absence of the pump beam (see Fig. 1).

To compare the performance of different cells, we use the gain coefficient,  $\Gamma$ , defined as  $\Gamma = (1/L) \ln[Gm/(m-G+1)]$ , where  $m$  is the incident beams intensity ratio. The dependencies of the coefficient,  $\Gamma$ , on the dc voltage,  $U$ , measured for the pure LC and for the colloid are presented in Fig. 2 for  $\Lambda \approx 11 \mu\text{m}$ . Ferroelectric particles do not change the character of the dependence  $G(V_{\text{dc}})$  but strongly enhance the energy exchange between the beams. At the optimum value of  $V_{\text{dc}} = 10$  V, the gain  $|\Gamma| \approx 100 \text{ cm}^{-1}$  for pure LC and  $|\Gamma| \approx 480 \text{ cm}^{-1}$  for the colloid. We found that an increase of the grating period resulted in an increase of the two-beam coupling gain both for the pure LC and for the suspension.

To understand the origin of the strong impact of ferroelectric particles on the beam coupling, let us consider which characteristics of the LC affect the beam coupling. As was established before [19], the beam coupling in the cells with PVK aligning layers is caused by light-induced spatial modulation of the surface charge layers. This modulation causes the appearance of modulated components of the electric field in the azimuthal and polar planes of the cell and, in turn, spatially modulated elastic torque. It results in spatially modulated reorientation of the director in the cell and in modulation of the refractive index that leads to the recording of the hologram and beam coupling. For small director deviation from the planar alignment [ $\theta(x, z) \ll 1$ ] spatial modulation of the refractive index can be expressed as [29]:

$$\tilde{n}_e(\psi, x, z) = \frac{n_e n_o}{(n_e^2 \sin^2 \beta + n_o^2 \cos^2 \beta)^{1/2}} - \frac{(n_e + n_o) \sin \beta \cos \beta}{(n_e^2 \sin^2 \beta + n_o^2 \cos^2 \beta)^{3/2}} n_a \theta(x, z), \quad (1)$$

where  $n_e$  and  $n_o$  are the main magnitudes of the refractive indexes along and perpendicular to the director (optical axis),  $n_a = n_e - n_o$  is the birefringence,  $\psi$  is the angle between director and light beam wave vectors, and  $\beta$  is the angle between cell normal and light beams.

Efficiency of the beam coupling depends on the amplitude of the refractive index modulation [second term in Eq. (1)] that is proportional to birefringence,  $n_a$ , and di-

rector distortion,  $\theta(x, z)$ . The distortion  $\theta(x, z)$  is determined by the minimum of the total free energy of a LC colloid. For the planar alignment of nematic in a cell and its strong anchoring with aligning surfaces, the total free energy in our geometry has the form [26]

$$F = \int_0^L \left\{ \frac{1}{2} (K_1 \cos^2 \theta + K_3 \sin^2 \theta) \theta'^2 - \frac{1}{2} \epsilon_0 (\epsilon_{\perp}^{\text{LC}} + \tilde{\epsilon}_a \sin^2 \theta) E_z^2(\theta) \right\} dz + \text{const}, \quad (2)$$

$$\tilde{\epsilon}_a^{\text{col}} = \epsilon_a^{\text{LC}} + c_{\text{part}} P^2 V \beta \lambda^2 / \epsilon_0, \quad (3)$$

where  $\tilde{\epsilon}_a^{\text{col}}$  is the effective dielectric anisotropy of the colloid,  $\epsilon_a^{\text{LC}}$  is the dielectric anisotropy of pure LC,  $\epsilon_{\perp}^{\text{LC}}$  is the component of the dielectric constant of pure LC perpendicular to the director,  $P$  is the permanent polarization of a particle,  $V$  is the particle volume,  $\lambda \sim 1$  is the local field correction factor, and  $K_1$  and  $K_3$  are elastic constants of the LC.

The enhancement of the orientational interaction in the colloid due to the particles was determined by comparing the values of  $T_c$ ; in the cells with a pure LC and those within the colloid. The clearing temperature,  $T_c$ , was determined by observing the LC textures in a polarization microscope at heating and cooling of the samples at a rate  $0.1 \text{ K min}^{-1}$ . We obtained the values  $T_c^{\text{LC}} \approx 58 \text{ }^\circ\text{C}$  for a pure LC and  $T_c^{\text{col}} \approx 70 \text{ }^\circ\text{C}$  for the colloid. Increasing of  $T_c$  in the colloid implies an increase of the orientational intermolecular interaction.

According to standard Maier–Saupe molecular field theory [30], the average order parameter of doped nematic,  $S(\tau)$ , is the universal function of the reduced temperature  $\tau = T/T_c$  and coincides with the dependence  $S_0(\tau)$  of a pure LC [31,32]. Therefore, expanding the dependence  $S^{\text{col}}(\tau^{\text{col}})$  into a power series  $\Delta\tau$ , we obtain

$$S^{\text{col}}(\tau^{\text{col}}) \approx S^{\text{LC}}(\tau^{\text{LC}}) + \Delta\tau \frac{dS^{\text{LC}}}{d\tau^{\text{LC}}}. \quad (4)$$

The shift of the reduced temperature is  $\Delta\tau = -[T/(T_c^{\text{LC}})^2] \Delta T_c \approx 3 \times 10^{-2}$  in our case, and the typical value of the derivative  $dS^{\text{LC}}/d\tau^{\text{LC}} \sim 1$  (far from the clearing temperature). Therefore, we can suggest that the increased order parameter in the colloid is negligibly small, and the enhancement of the beam coupling is not deter-

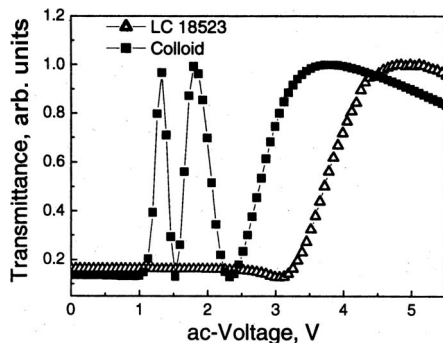


Fig. 3. Dependence of the transmittance of the cell on ac voltage measured for the LC and the colloid.

mined by the increased ordering of the nematic matrix due to the particles being presence.

To find changes of the birefringence of the LC due to the particles, we measured, the transmittance-voltage (TV) characteristics of the cells in the Freedericksz geometry. The cells with the pure LC and with the colloid were put between crossed polarizers so that the director made an angle of  $45^\circ$  with respect to the axes of the polarizers. The intensity of the probe beam of the He–Ne laser ( $\lambda = 0.63 \mu\text{m}$ ) was measured before and behind polarizers with increasing, applied ac voltage ( $\nu = 1 \text{ kHz}$ ). The obtained TV dependence (Fig. 3) is the result of a change in the LC orientation from planar to homeotropic. The number of extremes,  $m$ , is determined by the birefringence of the cell:  $m = n_a L / \lambda$ . We found at least three maxima in the cell with the colloid and only one maximum in the cell with a pure LC. It clearly demonstrates the increased birefringence of the colloid. Estimates show that  $n_a^{\text{LC}} \approx 0.05$  whereas  $n_a^{\text{col}} \approx 0.11$ . Since the change of the order parameter  $S$  in the colloid is small, we suggest that increased birefringence is caused by high-polarizability anisotropy of the ferrofluids at optical frequencies.

In the next stage of our investigation, we studied the dielectric anisotropy in the colloid and in the pure LC. The measurements were carried out by a standard bridge method at  $\nu = 1 \text{ kHz}$  in the cells with a planar alignment ( $\epsilon_{\perp}$ ) and homeotropic alignment ( $\epsilon_{\parallel}$ ) of LC. We obtained an increase of both components  $\epsilon$ :  $\epsilon_{\parallel}^{\text{LC}} = 7.2$ ;  $\epsilon_{\parallel}^{\text{col}} = 12.7$ ;  $\epsilon_{\perp}^{\text{LC}} = 4.2$ ;  $\epsilon_{\perp}^{\text{col}} = 5.2$ . Since the change of  $S$  in the colloid is small, the strong increase of the dielectric anisotropy at low frequencies ( $\epsilon_a^{\text{LC}} = 3.0$  and  $\epsilon_a^{\text{col}} = 7.5$ ) is determined by the direct contribution from the particles [see Eq. (3)] due to their ferroelectricity.

Furthermore, the increase of the dielectric anisotropy in the colloid should decrease the Freedericksz transition threshold. Freedericksz threshold voltage in the colloid is expressed as [26]

$$V_{\text{thr}}^{\text{col}} = \pi \sqrt{\frac{K^{\text{LC}}}{\epsilon_0 \tilde{\epsilon}_a^{\text{col}}}}. \quad (5)$$

It is assumed here that the dielectric anisotropy of the nematic matrix in the colloid is the same as of a pure LC, and  $K^{\text{col}} = K^{\text{LC}}$ . In our study of the particular system composed from LC18523 and  $\text{Sn}_2\text{P}_2\text{S}_6$  ferroelectric nanoparticles, there is no significant change in the order parameter  $S$ ; Frank elastic constants are proportional to  $S^2$  [33]; therefore, we assume that doping our LC with  $\text{Sn}_2\text{P}_2\text{S}_6$  ferroelectric nanoparticles does not affect the elastic constants.

The TV curves depicted in Fig. 3 demonstrate the evident decrease of the Freedericksz threshold voltage in the colloid:  $V_{\text{thr}}^{\text{LC}}/V_{\text{thr}}^{\text{col}} \approx 2.2$ . This ratio corresponds to the gain  $\tilde{\epsilon}_a^{\text{col}}/\epsilon_a^{\text{LC}} \approx 4.8$ . However, this value is more than two times larger than the one obtained from the direct measurements of dielectric anisotropy. This discrepancy and its origin need further investigation. One possibility is the decrease of the anchoring energy of the LC colloid that can lead to a lower Freedericksz transition voltage. Alternatively, the Frank constant in the colloid could be lower than in the pure LC, although there are no evident physical reasons for this effect.

### 3. CONCLUSIONS

We found the strong enhancement of two-beam coupling in the ferroelectric nanoparticles and LC colloid. The effect originated from an increased birefringence of the colloid and a stronger LC reorientation torque. Our measurements allowed us to suggest that increased birefringence is caused by the contribution of polarizability anisotropy of the ferroelectric particles. Stronger reorientation torque is caused by the permanent dipole moment of the particles contributing to the dielectric anisotropy of the colloid  $\epsilon_a^{\text{col}}$ . The enhancement of two-beam coupling in LCs by doping with ferroelectric nanoparticles at an extremely small concentration shows the strong potential of ferroelectric nanoparticles for improving the optical response of LCs, especially for those materials where a method of chemical synthesis has reached its limit. In our case, doping of the LC18523 with particles  $\text{Sn}_2\text{P}_2\text{S}_6$  leads to more than a twofold increase in the values  $n_a$  and  $\epsilon_a$ , and raised  $T_c$  by 12 °C as well as decreased  $V_{\text{thr}}$  by half. Colloids of ferroelectric nanoparticles in LCs clearly need to be investigated further to explore the full potential of those novel hybrid organic–inorganic materials in more detail.

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