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Sarik R. Nersisyan[a]; Nelson V. Tabiryan[a]
[a] Beam Engineering for Advanced Measurements Co., Winter Park, USA

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Microvibration-enhanced transparency of electrically aligned liquid crystals in nanoparticle networks

SARIK R. NERSISYAN and NELSON V. TABIRYAN*
Beam Engineering for Advanced Measurements Co., 809 S. Orlando Ave., Suite I, Winter Park, FL 32789, USA

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We study the process of orienting nanoparticle dispersed liquid crystals (NPD-LCs) with the aid of low-frequency (1–100 Hz) shearing microvibrations (1–10 μm amplitude). Due to the random orientation of LCs in such networks, NPD-LCs are highly light scattering unless subject to the orienting influence of a high-voltage electric field (~100 V). We show that microvibrations are able considerably to enhance the stable NPD-LC transmission state obtained due to the application of an electric field. Shearing microvibrations make it feasible to switch between the transparent and opaque states of the material in an all-mechanical process. The optical transmission of the material proves to be very sensitive to the vibration amplitude as well as frequency. As a result, the system can be used for sensing and measuring low-frequency and small-amplitude mechanical oscillations.

1. Introduction

Nanoparticle dispersed liquid crystals (NPD-LCs) have been studied for many years due to their interesting fundamental properties and promising applications [1–9]. These materials are made by dissolving a small amount of, particularly, silica nanopowder in a LC. At a certain concentration, silica particles are freely suspended in the solvent LC. With an increase in concentration of the particles, they start to aggregate, forming a fragile network. The fragility of the network is due to the weakness of the hydrogen bonding in the silica matrix, 4–40 kJ mol^{-1}, which is, however, still stronger than van der Waals forces.

The nanoparticle network splits the volume of the LC into randomly oriented regions of microscopic sizes. The system of randomly oriented, highly anisotropic microdomains exhibits strong light scattering and results in an opaque material. Due to the fragility of the nanoparticle network, the application of an electric field is capable of orienting the whole volume of the LC along the electric field direction, thus practically eliminating light scattering [5, 7]. Both the scattering and the transparent states of the material are stable due to relative rigidity of the network and its adaptation to the state of orientation.

In this paper, we present the results of detailed characterization of the previously reported [10] effect of microvibrations on NPD-LCs. We show that the combination of microvibrations with electric fields switches the material into the state of highest optical transmission, which is preserved upon removal of the external influences.

2. NPD-LC orientation due to shearing microvibrations

In this section, we characterize the effect of microvibrations on a NPD-LC, as reported by us previously [10]. The experimental set-up is shown schematically in figure 1. The material used in these tests was prepared using LC E48 (Merck) doped by 3 wt % of aerosil R974 (Degussa) with 12 nm particle size. One of the substrates of the cell containing the NPD-LC is fixed on a base, whereas the second substrate is connected to a piezo-actuator that is brought into an oscillatory motion by application of an a.c. electric field from a function generator. The effects under discussion were pronounced for voltages ~10 V which corresponded to ~1 μm amplitude of vibrations. The transmission state of the LC was monitored using a laser beam of 1.06 μm wavelength.

Figure 2 a shows the change in transmission of an $L=10\,\mu\mathrm{m}$ thick NPD-LC cell for different values of voltages applied to the piezoactuator at the frequency $\Omega=10$ Hz. The transmission of the same cell for fixed amplitude of the electric voltage at different frequencies is shown in figure 2 b.

Increasing the material thickness leads to exponential decrease of the transmission of the opaque state of the NPD-LC. Provided we are able to reach the state of

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*Corresponding author. Email: nelson@beamco.com

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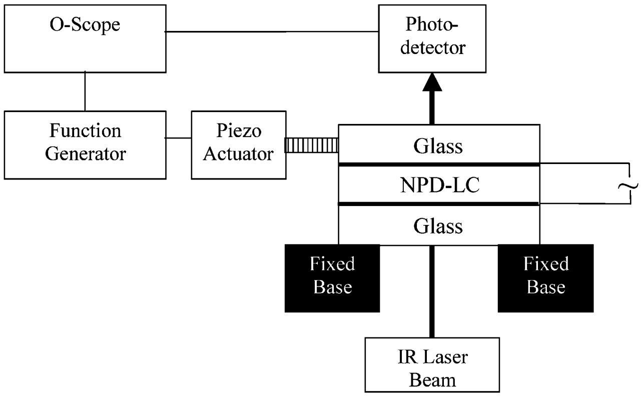

Figure 1. Schematic of the experimental set-up for studying the effect of shearing on optical and electro-optical properties of NPD-LCs.

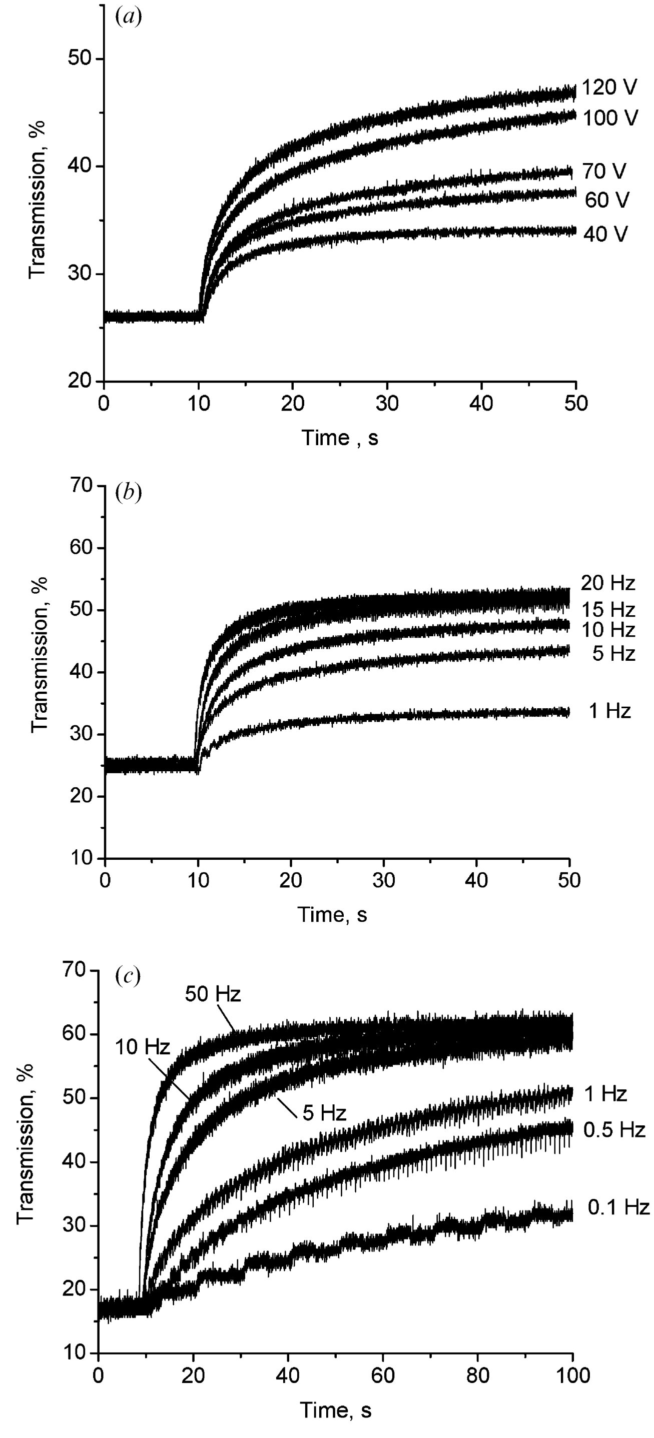

Figure 2. Vibration-induced increase in the transmission of NPD-LC cells: (a) L=10 µm, 40 V<V<120 V, f=10 Hz sinusoidal modulation; (b) L=10 µm, V=120 V, 1<f<20 Hz sinusoidal modulation; (c) L=20 µm, V=120 V, 0.1<f<50 Hz rectangular modulation. The NPD-LC consists of LC E48 (Merck) doped by 3 wt % aerosil R974 (Degussa).

high transmission in such a cell, we shall be able to increase exponentially the contrast of switching between the two optical states of the material by increasing the layer thickness. We performed tests on vibration-induced transparency in thick layers using a 50 µm thick cell of the material LC E48 doped with 3 wt % aerosil R974. Even though microvibrations could increase the transmission of 50 µm thick NPD-LC, the obtained levels were smaller than in thinner cells, due, apparently, to the fact that the vibrations did not propagate far enough, remaining localized near the surface of the shearing substrate.

Plots of the contrast ratio as a function of voltage at fixed frequency values and as a function of frequency at fixed voltage values are shown in figure 3 for cells of different thickness. The graphs reveal that microvibrations affect the thinner cell more efficiently, and that rectangular pulses of the electric field result in the fastest acceleration of the piezoactuator to its maximum extension value, as determined by the amplitude of the voltage (see also figure 2 c).

The enhanced transparency of the material is a result of LC orientation in the nanoparticulate network, which was verified under crossed polarizers. Such an orientation along the direction of shearing vibrations could be expected in the light of the well-known property of polymer network LCs to align along the direction of shearing deformations [11]

3. Combination of mechanical and electrical influences

Microvibrations are capable of increasing the NPD-LC transmission to an intermediate value of about 50%. Application of an a.c. electric field (typically 1 kHz frequency) makes it possible to reach transmission values close to 100%. However, different sequences and

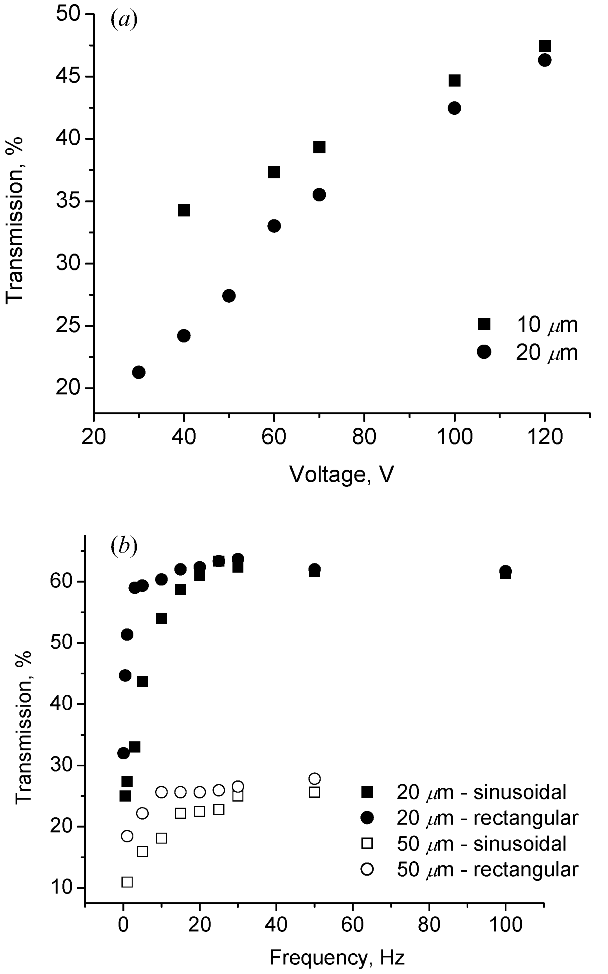

Figure 3. (a) Transmission as a function of voltage for 10 and 20 μm thick layers of NPD-LC subject to microvibrations at 10 Hz for 50 s. (b) Transmission as a function of frequency for 20 and 50 μm thick NPD-LC layers subject to sinusoidal and rectangular microvibrations for 100 s at 120 V piezo-actuator drive voltage.

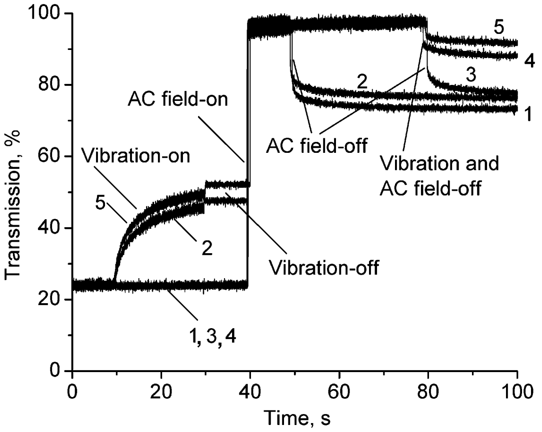

Figure 4. Transmission of a 10 μm NPD-LC as a function of time for different combination regimes of microvibrations and electrical field: 1, electric field on for 10 s; 2, electric field on for 10 s following vibrations; 3, electric field on for 40 s; 4, vibrations following application of the electric field for 30 s; 5, vibrations performed both before and after application of the electric field. The amplitude of the electrical voltage applied to the cell is 100 V at 1 kHz. The piezoactuator is driven by a sinusoidal electrical voltage of 120 V and frequency of 10 Hz.

combinations of microvibrations and the electric field lead to rather different final results.

The effect of different combinations of the mechanical and electrical influences on 10 μm thick NPD-LC is shown in figure 4. The NPD-LC transmission in the state of high transparency is practically the same and close to 90% in the presence of an electric voltage of ~100 V, whether or not the cell has previously been processed by microvibrations (see curves 1 and 2 in figure 4). Thus, microvibrations applied before the application of an electric field do not affect the level of electrically induced transparency of the NPD-LC. The state of maximum transmission obtained in an electric field relaxes to a considerably lower level when the electric field is switched off. The resultant field-off transmission level is slightly higher if the field is applied after subjecting the cell to microvibrations (compare curves 1 and 2 in figure 4).

Application of the electric field for extended periods of time does not improve appreciably the resulting field-off level of the cell transmission (see curve 3 in figure 4). Considerable improvement of the field-off transmission level can be obtained, however, if the microvibration procedure is performed while the electric field is on (see curves 4 and 5 in figure 4). The highest transmission level is obtained when microvibrations are performed both before *and* during the applications of the electric field (see curve 5 in figure 4).

Performing microvibrations after switching off the electric field may have different consequences depending on the electric field strength. If the transparent state is obtained by application of a relatively small voltage, microvibrations lead to further increase in the NPD-LC transmission, whereas the transparent state deteriorates if it was created by voltage of around 100 V (see figure 5).

Thus, we can conclude that the state of highest transmission of a NPD-LC layer can be obtained by subjecting the cell to shearing microvibrations before and during the application of an electric voltage. The transparency of the field-off state increases with applied voltage (see figure 6). It is important to stress that microvibrations should be performed while the electric field is on. If the electric field is switched off, subjecting the cell to microvibrations does not improve and might

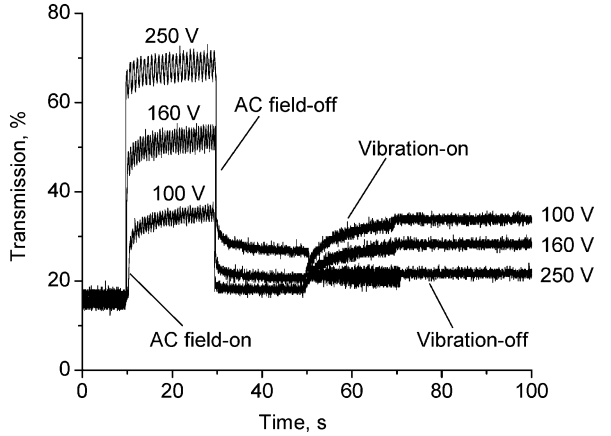

Figure 5. The effect of microvibrations on the transmission of 20 μm thick layer of NPD-LC performed after switching off the electric field.

even degrade the state of transparency established after switching off the electric field.

4. Mechanical restoration of the opaque state

The opaque state of strong light scattering can be restored mechanically by inducing shear over a relatively large distance of about 50 μm in one direction. The process of mechanical restoration of the strong light scattering state is quite fast, about 150 ms in a 10 μm thick NPD-LC and about 200 ms in a 20 μm thick NPD-LC, if the transparent state is obtained due to the combined action of microvibrations and an electric field (see figure 7). Thus, we have a very efficient all-mechanical system for switching between different optical states of the material.

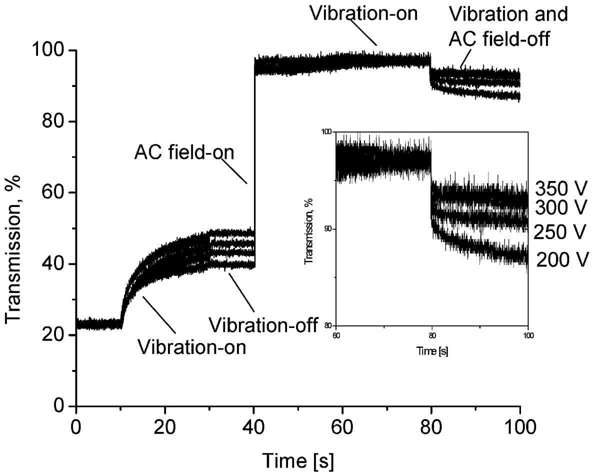

Figure 6. Transmission as a function of time for different voltages applied to a 10 μm thick NPD-LC in the optimum regime of combination of mechanical and electrical influences on the cell. Microvibrations are induced at 10 Hz by applying a 120 V sinusoidal electrical signal to the piezoactuator.

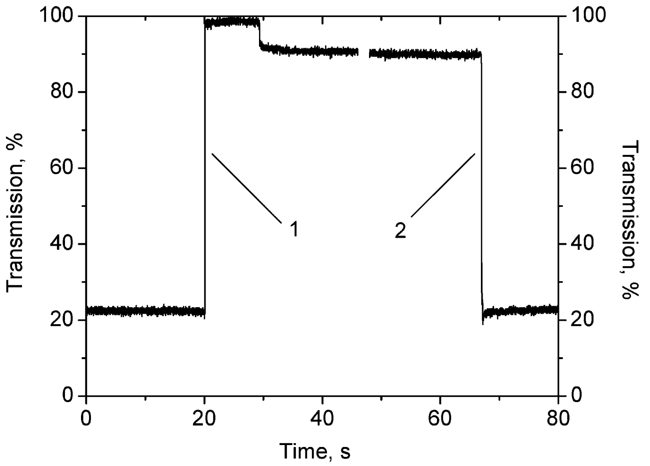

Figure 7. Mechanical (manual) restoration of the high-scattering state of a NPD-LC of thickness 10 μm due to extended shearing of over 50 μm within 1 s with the aid of a micrometer.

5. Discussion

The material under discussion is a complex system due to the many 'degrees of freedom' related to both the LC and the nanoparticulate network. Only a qualitative description of their electro-optical properties is so far available [7, 12]. The development of a theoretical model that could quantitatively describe the results of our observations presents an even greater challenge. At this stage of our studies we can therefore only present an imprecise picture of the processes taking place in the material.

The basis of such a picture is the thixotropic property of aerosils, well known and used in many of their applications [13–17]. Shearing microvibrations disrupt the network, dramatically reducing its viscosity. Flow gradients orient the long hydrogen-bonded nanoparticle chains [18], thus orienting the LC molecules. When the vibrations are switched off, the nanoparticle network is restored, maintaining its orientation and hence the orientation of the LC molecules. The time scale of the network restoration is known to be about 1 s, which is longer than the typical relaxation time of a LC in small confinements.

The simplest expression that may be considered to fit the obtained curves should proceed from exponential relaxation of the coefficient, α, characterizing the extinction of radiation intensity, I, in a NPD-LC layer of thickness L due to light scattering, i.e. $I \sim \exp(-\alpha L)$. We shall take into account that, in general, the process is described by relaxation times τ_1 and τ_2 related to the

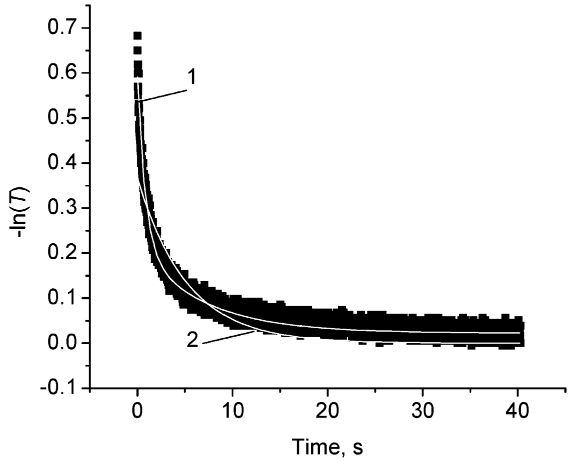

Figure 8. A typical relaxation process and its fit with a single exponential decay function (1) and with two exponential decay functions (2).

LC orientation and network reconfiguration. Thus, the dynamics of transmission T of the material can be written as

$$-\ln T = \alpha L = A_0 + A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2). \quad (1)$$

Figure 8 shows an example of a fit using equation (1). The magnitudes of the time constants in this case were $\tau_1 = 0.8$ s and $\tau_2 = 6.9$ s. Apparently, the longer time scale describes the reconstruction and alignment of the nanoparticle network, whereas the shorter time scale is related to orientation of the LC accommodating these changes. Statistical processing of data yields $\chi^2 = 0.001$ and $R^2 = 0.8$ for the single exponential fit, and $\chi^2 = 0.00019$ and $R^2 = 0.97$ for a fit using two exponential functions.

The slope of the linear initial stage of transmission change with time increases with increasing voltage and frequency, as shown in figure 9 for 10 and 20 µm thick NPD-LCs. The 10 µm thick cell reveals considerably higher sensitivity than the 20 µm thick cell studied in our earlier work [10].

6. Conclusions

We have characterized the possibility of switching between the transparent and opaque states of NPD-LC material systems in an all-mechanical process that can be performed without involving electrical fields. The combination of microvibrations and electric fields allows considerable improvement of the high-transmission state of a NPD-LC compared with the influence of an electric field alone. The state of highest transmission of a NPD-LC layer is obtained by subjecting the cell to shearing microvibrations before and during the application of an electric voltage. If the electric field is switched off, subjecting the cell to microvibrations does not improve and might even degrade the state of transparency established after switching off the electric field.

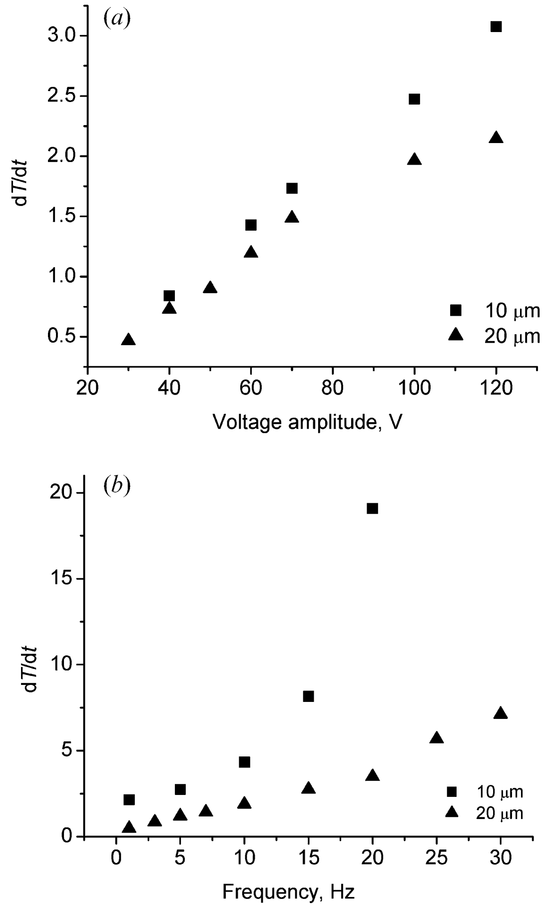

Figure 9. Slope of the linear region of transmission dynamics as a function of (a) the amplitude of the sinusoidal electrical voltage driving the piezoactuator at 10 Hz and (b) the frequency of the sinusoidal voltage and 120 V amplitude. Data are shown for 10 and 20 µm thick NPD-LC cells.

The optical transmission of the material proves to be very sensitive to the vibration amplitude as well as frequency and, as a result, the system can be used for sensing and measuring low-frequency and small-amplitude mechanical oscillations.

Acknowledgement

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