Influence of DC voltage on the dielectric properties of nematics

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Abstract. We report on precise impedance measurements, with the aim of exploring the influence of a dc bias voltage on the dielectric permittivity and electrical conductivity of liquid crystals and on their anisotropies. We prove that the dielectric permittivity is not affected by a dc bias; however, the electrical conductivity suffers a substantial reduction upon increasing the superposed dc voltage. Moreover, we show that the relative conductivity anisotropy also diminishes at increasing dc bias.

Introduction

Nematic liquid crystals are anisotropic dielectric materials with a low electrical conductivity, which originates from a small concentration of ionic contaminants. Experimental studies of the electro-optical properties of nematics, as well as most display applications of them utilize ac driving voltage; investigations under dc voltage are rather scarce. Recently, electroconvection and flexoelectric patterns occurring under superposed action of ac and dc voltages have been studied and, unexpectedly, an inhibition of the pattern formation at high voltages (an increase of the thresholds) was found [1-2]. The fact that conductivity is an important factor determining the threshold voltage, motivated the present testing of the effect of dc bias on the electrical conductivity.

Experimental Method

Measurements were performed on the compound 4-n-octyl-oxyphenyl 4-n-methyloxybenzoate (1OO8), having a nematic phase in the temperature range between 53 ^{o}C and 77 ^{o}C and a chemical structure shown in Fig. 1. It is a material used for studying pattern formation in electric field [2-3]. Precise impedance measurements were carried out with the dielectric analyzer Novocontrol Alpha equipped with a ZG4 test interface. The liquid crystal was filled into a plane condenser (glass plates with transparent electrodes of $A = 1$ cm^{2} size, separated with a gap of $d = 1$ mm). The electrodes were coated with polyimide layer, in order to mimic the electrical boundary conditions of the thinner commercial cells employed in pattern formation studies. The cell was placed in a shielded chamber, with its temperature kept at $T = 58 \pm 0.05$ ^{o}C.
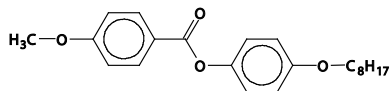

Fig. 1. The chemical structure of the nematic 4-n-octyloxyphenyl 4-n-methyloxybenzoate (1OO8).

As liquid crystals are anisotropic materials, the dielectric permittivity (electrical conductivity) can be characterized by the quantities $\varepsilon_{\parallel}$ ($\sigma_{\parallel}$) and $\varepsilon_{\perp}$ ($\sigma_{\perp}$), measured with an electric field **E** parallel with and perpendicular to the director **n**, respectively. These two orthogonal experimental geometries can be easily realized, if the chamber is put into a magnetic field **H**. For the large cell thickness used, a magnetic induction value of $B = 1$ T is large enough to align the director parallel to **H**. Thus applying

a magnetic field parallel to the electrode surface ($\mathbf{H}, \mathbf{n} \perp \mathbf{E}$), the perpendicular component is obtainable; when the chamber is rotated so that the magnetic field becomes normal to the electrodes ($\mathbf{H}, \mathbf{n} \parallel \mathbf{E}$), the parallel component can be measured.

The complex impedances of the empty and the liquid- crystal-containing cell were measured at the frequency of 1 kHz, with a probing ac rms voltage of $U_{ac} = 0.2$ V. The liquid crystal sample was interpreted as a parallel RC circuit, with

$$R_i = \left(\sigma_i \frac{A}{d} \right)^{-1} ; C_i = \varepsilon_0\, \varepsilon_i \frac{A}{d}, \text{ where } i = \parallel, \perp . \qquad (1)$$

The dc bias voltage was incremented from $U_{dc} = 0$ up to $U_{dc} = 40$ V in 1 V steps, in each 10 minutes. At a fixed bias voltage, measurements were performed repeatedly several times, alternately in the parallel and perpendicular geometries. Permittivity and conductivity values were evaluated using (1).

Results

Measurements proved that the dielectric permittivity values do not depend on the dc bias voltage: we obtained $\varepsilon_\parallel = 4.96 \pm 0.01$ and $\varepsilon_\perp = 5.45 \pm 0.01$; i.e., the dielectric anisotropy, $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$, is negative, as expected from the molecular structure in Fig. 1. The small deviations of about 0.2% are attributed to temperature fluctuations. The constancy of $\varepsilon_\parallel$ and $\varepsilon_\perp$ indicate that the dc bias did not alter the director alignment created by the magnetic field; i.e., no pattern formation occurred at the applied voltages.

In contrast to the permittivity, the electrical conductivity substantially depends on the bias voltage. Figure 2(a) exhibits the dc bias dependence of $\sigma_\parallel$ and $\sigma_\perp$, as well as that of the conductivity anisotropy $\sigma_a = \sigma_\parallel - \sigma_\perp$; the symbols show the values averaged over a 10 minutes interval. It is clearly seen that the bias dependence is nonlinear. For $U_{dc} < 4$ V the change is small, there is even a slight increase of σ; however, for $U_{dc} > 4$ V, increasing the dc bias reduces strongly the conductivities. The maximal reduction is by about a factor of 6. The conductivity anisotropy also diminishes, though remains positive in the whole tested voltage range. It is important to note that the relative conductivity anisotropy, defined as $\delta\sigma = \sigma_a / \sigma_\perp$, is decreasing with the dc bias voltage too, as depicted in Fig. 2(b); at the highest bias it is only about 60% of its initial value at $U_{dc} = 0$.

The temporal dynamics of how the conductivity responds to voltage variations was also tested. Unsurprisingly, σ does not change instantaneously after a switching on/off the dc bias voltage; the voltage jump initiates a relaxation process instead. Measurements proved that this relaxation is rather long; moreover, it can be characterized by several relaxation times, from seconds up to several hours, indicating the presence of various ionic processes (e.g., migration, diffusion, association, recombination, and adsorption).

Discussion

By its original concept, a constant electrical conductivity describes the linear, voltage independent current response of the system to the electric excitation; i.e., it implicitly assumes ohmic conductivity. Though in liquid crystals conductivity is mostly due to ions, under ac voltage the assumption of ohmic conductivity is usually an acceptable approximation. This fails, however, in the presence of dc voltage. A dc voltage induces migration of ions leading to a partially ion-depleted region in the bulk, the formation of a charged double layer with high electric field gradients near the electrodes, and adsorption of ions at the substrates. It is not surprising that these phenomena result in a reduction of the number of active charge carriers and thus in dc voltage dependent, diminishing electrical conductivity. On the other hand, the relative conductivity anisotropy is assumed to depend rather on the structure of the nematic matrix, which is not expected to change with the number of charge carriers. Thus the dc- voltage-induced reduction of the relative conductivity anisotropy is an unexpected founding; it may perhaps be related with a change in the type of charge carriers.

Electrical conductivity, as well as its (relative) anisotropy is a key, though hardly controllable material parameter for governing the formation of electroconvection patterns. Usual theoretical

modelling of electroconvection phenomena assumes ohmic conductivity with constant $\sigma_\parallel$ and $\sigma_\perp$. The results shown above clearly demonstrate that this assumption fails and should be given up if the applied voltage has a dc component. Therefore, if it is not done so, a mismatch between theoretical predictions and experimental observations about the onset characteristics (threshold voltage, critical wave number) of electroconvection patterns is practically unavoidable and actually has been reported for patterns induced by superposed ac and dc voltages [1-2]. A precise description of all ionic processes mentioned above and thus the understanding of the dc-voltage-induced conductivity changes would be a huge challenge for the future development of the theory of electric-field-induced pattern formation.
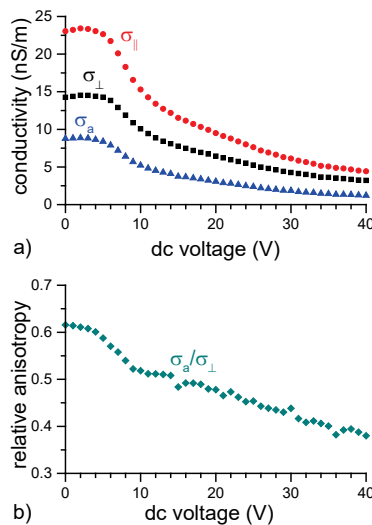

Fig. 2. Dc voltage dependence of a) the electrical conductivity parallel with ($\sigma_\parallel$) and perpendicular to ($\sigma_\perp$) the director, and the anisotropy $\sigma_a = \sigma_\parallel - \sigma_\perp$; b) the relative anisotropy $\sigma_a/\sigma_\perp$.

Some qualitative features of the pattern formation at superposed ac and dc voltages can, however, be estimated by testing the effect of varying $\sigma_\perp$ and $\delta\sigma$ on the pattern onset voltages. Recent simulations have shown that reduction of $\delta\sigma$, that occurs upon increasing the dc bias voltage, results in the increase of the threshold voltage of electroconvection, independently of the frequency of the ac voltage component [2]. This offers an explanation for the experimental observations and thus resolves the mismatch with theory.

Acknowledgment

References
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