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G. Barbero[ab]; A. M. Figueiredo Neto[a]; J. Le Digabel[ac]; O. G. Martins[a]
[a] Instituto de Física, Universidade de São Paulo, São Paulo, SP - Brazil [b] Dipartimento di Fisica del Politecnico, Corso Duca degli Abruzzi 24, 10129 Torino, Italia [c] Ecole Normale Superieure de Cachan, 61 Av. du President Wilson, 94235 Cachan Cedex, France

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Ionic relaxation in nematic liquid crystal cells

G. BARBERO†‡, A.M. FIGUEIREDO NETO†, J. LE DIGABEL‡§ and O.G. MARTINS†
†Instituto de Física, Universidade de São Paulo, P.O. Box 66318, São Paulo, 05389-970, SP - Brazil
‡Dipartimento di Fisica del Politecnico, Corso Duca degli Abruzzi 24, 10129 Torino, Italia
§Ecole Normale Superieure de Cachan, 61 Av. du President Wilson, 94235 Cachan Cedex, France

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We investigate the dependence of the relaxation time of the current flowing in a nematic cell submitted to an external dc voltage on the physical properties of the substrate. We show that previously presented analyses of the same problem are not very useful for practical applications. We compare our theoretical predictions with experimental data, and show that the agreement is rather good. The influence of the adsorption–desorption phenomenon on the relaxation time is also discussed.

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1. Introduction

Thermotropic nematic liquid crystals are organic materials whose electrical conductivity is of ionic origin [1]. When they are submitted to an alternative external voltage of a few kHz, they can be considered as insulating media because the ions do not follow the rapid variations of the applied electric field [2]. On the contrary, if the external field is dc or of low frequency, the presence of the ions strongly modify the optical response of the nematic sample [3]. The ions are responsible for a surface electric field that renormalizes the effective surface energy [4]. Furthermore, in the presence of a low-frequency or dc field, the ions are responsible for a strong reduction of the electric field in the sample [5]. The importance of the ions on the optical properties of a nematic cell submitted to a dc, or low-frequency, electric field has been considered recently [6, 7]. Of course, the presence of the ions is also responsible for the relaxation of the electrical current in the circuit containing the nematic cell submitted to a step-like external voltage. This problem has been considered in detail by several authors, by describing the nematic cell by means of equivalent circuits [8–10]. In this paper we want to reconsider this problem and to show that the circuital analysis is not very useful to determine the relaxation time of the current. In section 2 we present the standard circuital analysis and underline its weak point. In section 3 we determine the relaxation time by considering the drift-diffusion of the ions in the presence of an external field. In section 4 we compare our predictions with the experimental data reported by Sieberle and Schadt [8]. The influence of the adsorption–desorption phenomenon on the relaxation time is discussed in section 5. Section 6 is devoted to the conclusions.

2. Circuital analysis

We assume that the media involved in the description are characterized, from the electrical point of view, by a dielectric constant, ε, and a conductivity, σ. The sample is in the shape of a slab of thickness d. The surface layer, deposited on the electrodes to orient the nematic liquid crystal, has a thickness δ. We indicate by the subscripts S and B the properties relevant to the surface layer and to the bulk, respectively, and by: (i) V, the potential applied to the sample; (ii) Σ_i, the surface charge density accumulated at the interface between the nematic liquid crystal and the surface layer; (iii) Σ_e, the surface charge density sent by the power supply on the electrodes to impose the potential V; (iv) I, I_S, and I_B, the electrical current in the external circuit, in the surface layer and in the bulk of the sample, respectively. The applied voltage is supposedly so low that the chemical equilibrium relevant to the dissociation of the impurity producing the ions is not perturbed by the presence of the external electric field [11]. In the quasi-static case, from the Maxwell equation

$$\nabla\times\mathrm{B}=\mu_0\left(J+\frac{\partial \mathrm{D}}{\partial t}\right), \tag{1}$$

where the meaning of the symbols is well known, we obtain

$$J=\frac{\mathrm{d}\Sigma_e}{\mathrm{d}t}+J_S, \quad \text{and} \quad J_S=-\frac{\mathrm{d}\Sigma_i}{\mathrm{d}t}+J_B, \tag{2}$$

where J, J_S and J_B are the current densities in the external circuit, in the surface layer and in the bulk, respectively. By definition we have $J_S=\sigma_S E_S$ and $J_B=\sigma_B E_B$, where E_S and E_B the electric field in the surface layer and in the bulk, respectively [6]. From the

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Maxwell equation, $\nabla\cdot\mathbf{D}=\rho$, a simple calculation gives

$$E_S=\frac{\Sigma_e}{\varepsilon_S},\quad \text{and} \quad E_B=\frac{\Sigma_e-\Sigma_i}{\varepsilon_B}. \tag{3}$$

Since the difference of electrical potential applied at the sample is V, it follows that 2 $E_S\delta+E_Bd=V$. From this conditions, and taking into account equation (3) we obtain

$$\Sigma_e=\gamma\left(\varepsilon_B\frac{V}{d}+\Sigma_i\right), \tag{4}$$

where $1/\gamma=1+2(\varepsilon_B\delta)/(\varepsilon_S d)$. By substituting equation (4) into equation (3) we get that the electric fields in the surface layer and in the bulk are given by

$$\begin{aligned} E_S&=\frac{\gamma}{\varepsilon_S}\left(\varepsilon_B\frac{V}{d}+\Sigma_i\right),\\ E_B&=\gamma\frac{1}{d}\left(V-2\frac{\delta}{\varepsilon_S}\Sigma_i\right). \end{aligned} \tag{5}$$

By substituting equations (5) into $J_S=\sigma_S E_S$ and $J_B=\sigma_B E_B$ and the result into equations (2) we get

$$\frac{\mathrm{d}\Sigma_i}{\mathrm{d}t}=\alpha\frac{V}{d}-\frac{1}{\tau}\Sigma_i, \tag{6}$$

where

$$\begin{aligned} \alpha&=\gamma\left(\sigma_B-\sigma_S\frac{\varepsilon_B}{\varepsilon_S}\right),\\ \frac{1}{\tau}&=\frac{\gamma}{\varepsilon_S}\left(\sigma_S+2\frac{\delta}{d}\sigma_B\right). \end{aligned} \tag{7}$$

As is clear from equation (6), τ is the relaxation time of the effect connected with the time evolution of the surface density of ions Σ_i. From equation (4) and equation (2), τ is also the relaxation time for the current in the external circuit. Since Σ_i is connected with the ionic charge accumulated at the interface with the surface substrate due to the current in the bulk, equation (6) has to be solved with the boundary condition $\Sigma_i(0)=0$. The relaxation time given by equation (7) can be written as

$$\tau=\frac{(\varepsilon_S/\delta)+2(\varepsilon_B/d)}{(\sigma_S/\delta)+2(\sigma_B/d)}. \tag{8}$$

Equation (8) when the substrate is a pure insulating material, and hence $\sigma_S=0$, reads

$$\tau=\frac{1}{2}\left(\frac{\varepsilon_S}{\delta}+2\frac{\varepsilon_B}{d}\right)\rho_B\, d, \tag{9}$$

where $\rho_B=1/\sigma_B$ is the resistivity of the nematic liquid crystal. Equation (8) coincides with the one obtained by Sieberle and Schadt [8]. Despite its apparent simplicity this equation is not useful for practical application for the following reason. In equation (8) ε_B and σ_B are phenomenological parameters [12]. It is not clear if they are relevant to the pure liquid crystal or to the liquid crystal containing ions. In this second case, how they depend on the ionic concentration is not evident [12]. Only after solving the complete problem of the diffusion of ions in the pure liquid crystal is it evident how the phenomenological parameters depend on the ions. In other words, the earlier analysis [8] was performed by approximating the nematic liquid crystal by an electrical circuit formed by a resistance and a capacitance in parallel, in agreement with equation (1). However, this circuital approximation in the dc limit does not work for a nematic liquid crystal, whose conductivity is of ionic origin [12]. For this reason, in the following section we reconsider the ionic relaxation from another point of view.

3. Drift-diffusion analysis

The rigorous analysis of the problem under consideration has to be performed by solving the equations of continuity for the positive and negative ions, and the Poisson equation for the electrical potential [13]. Assuming that there is no selective ion adsorption from the limiting surfaces, before the application of the external electric field the sample is locally and globally neutral. When the external field is applied, the positive ions move toward the negative electrode and the negative ions toward the positive electrode. In this situation the sample is still globally neutral, but locally charged. The time evolution of the electrical charge density in a given point of the sample depends on the mobilities of the ions, their electrical charge, and on the amplitude of the applied external voltage between the electrodes. This kind of analysis has been performed recently by Thornton *et al.* [14], in the limit of small applied voltage, where the basic equations of the problem can be linearized. Thornton *et al.* [14] show that the ionic relaxation is characterized by a typical relaxation time given by

$$\tau_{\text{ion}}=\frac{\lambda d}{2D}, \tag{10}$$

where $\lambda=\sqrt{\varepsilon KT/(2n_0q^2)}$ is the Debye length [15], d the thickness of the sample, assumed in the shape of a slab, and D the diffusion coefficient of the ions in the liquid. In the definition of λ reported above, ε is the dielectric constant of the pure liquid in which the ions are dispersed, KT the thermal agitation energy, n_0 the bulk density of ions, in thermodynamical equilibrium, and q the electrical charge of the ions. Equation (10) holds in the case in which the ions are identical in all the aspects,

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except for the sign of the electrical charge. In the opposite case equation (10) it is a little bit more complicated, but it maintains a similar form. Hence, according to Thornton *et al.* [14], the relaxation time of the ions in an electrolytic solution, in the low frequency case, is given by equation (10). For a commercial liquid crystal, $\lambda \sim 0.1\,\mu\mathrm{m}$ [16] and $D \sim 10^{-11}\,\mathrm{m}^2\,\mathrm{s}^{-1}$ [17]. Consequently, if $d = 10\,\mu\mathrm{m}$, one gets $\tau \sim 0.1\,\mathrm{s}$. On the contrary, for a purified liquid crystal, for which $\lambda \sim 1\,\mu\mathrm{m}$, the relaxation time for a cell having the same thickness is of the order of 1 s [1]. The relaxation time given by equation (10) has been deduced [14] by solving the fundamental equations of the problem by means of the Laplace transformation technique. More recently, it has been obtained by solving the same equations in the standard manner, in which the solutions of the problem are separated, in the linear approximation, in the transient and steady components [18]. The solution relevant to the transient components gives rise to an eigenvalues equation for the relaxation times. The smallest solution of this eigenvalues equation, having a physical meaning, is coincident with the one given by equation (10). The importance of the ions on the impedance spectroscopy has been considered by Barbero and Alexe-Ionescu [19], by generalizing the analysis proposed by Cirkle *et al.* [13]. The impedance of an insulating liquid containing ions was evaluated [19] without any circuital analysis, by solving the drift-diffusion problem for the ions, when the sample is submitted to a sinusoidal external potential of small amplitude, assuming that the electrodes are perfectly blocking. In the low-frequency limit [19], the impedance of the cell can be written as $\mathcal{Z} = \mathcal{R} - i\mathcal{X}$, where $\mathcal{R}$ is the resistance of the cell, and $\mathcal{X} = 1/(\omega C)$ its reactance. According to elementary electrodynamics,

$$\mathcal{R} = \frac{1}{\sigma_{\mathrm{eq}}}\frac{d}{S}, \quad \text{and} \quad C = \varepsilon_{\mathrm{eq}}\frac{S}{d}, \tag{11}$$

where σ_{eq} and $\varepsilon_{\mathrm{eq}}$ are the equivalent electrical conductivity and equivalent dielectric constant of the medium, renormalized by the presence of the ions dissolved in the liquid, S is the surface area of the electrodes, and $\omega = 2\pi f$ the angular frequency of the applied potential. In the limit $\omega \to 0$, as shown previously [19]

$$\sigma_{\mathrm{eq}} = 2 n_0 \mu q, \quad \text{and} \quad \varepsilon_{\mathrm{eq}} = \frac{1}{2}\varepsilon\frac{d}{\lambda}, \tag{12}$$

where μ is the mobility of the ions in the liquid. As for equation (10), equations (12) are valid when the ions have the same mobility. From equations (12), taking into account that $\lambda \ll d$, it follows that in the dc limit $\varepsilon_{\mathrm{eq}} \gg \varepsilon$, as it has been experimentally observed [20]. By using equations (12) the dielectric relaxation time [12] of the sample, defined by $\tau_{\mathrm{eq}} = \varepsilon_{\mathrm{eq}}/\sigma_{\mathrm{eq}}$, is found to be

$$\tau_{\mathrm{eq}} = \frac{\varepsilon d}{4\lambda \mu n_0 q}. \tag{13}$$

From equation (13), by taking into account the Einstein–Smolucowsky relation, $\mu/D = q/(KT)$ [21], and the definition of the Debye length, it is possible to eliminate n_0, and to show that $\tau_{\mathrm{eq}} = \tau_{\mathrm{ion}}$, where τ_{ion} is given by equation (10). It follows that equation (10) can be considered as the dielectric relaxation time of an insulating liquid containing ions. It is important to underline that, in the limit $\omega \to 0$, the nematic liquid crystal can be approximated by a series of an ideal condenser with a pure ohmic resistance, given by equation (11). The phenomenological parameters representing the conductivity and the dielectric constant, renormalized for the presence of the ions dissolved in the liquid crystal, are given by equation (12).

4. Comparison with the experimental data

Using the consideration reported above, we can now evaluate the influence of the surface treatment on the effective relaxation time of a nematic cell submitted to a step-like external field. To this aim we assume, as did Sieberle and Schadt [8], that the electrodes are covered with a plastic material, of thickness δ, whose dielectric constant is ε_S, that can be considered a perfect insulating material. This surface layer is important, from the technological point of view, because it allows orientation of the nematic liquid crystal. Furthermore, it avoids injection of charge in the nematic cell. In other words, when the electrodes are covered with the polymer, they can be considered as blocking. From the electrical point of view the nematic sample is a series of the condenser, of capacitance $C_S = \varepsilon_S(S/\delta)$, representing the surface treatments, with the nematic liquid crystal. By taking into account that, in the limit $\omega \to 0$, the nematic liquid crystal is equivalent to an R-C series, as discussed above, we have that the total capacitance of the cell is $C_T = C_S C/(2C + C_S)$, and the relaxation time is $\tau_r = RC_T$. By taking into account equation (12), τ_r can be rewritten as

$$\tau_r = \frac{1}{2}\frac{\rho d}{(\delta/\varepsilon_S) + (\lambda/\varepsilon)}, \tag{14}$$

where $\rho = 1/\sigma_{\mathrm{eq}} = 1/(2 n_0 \mu q)$ is the resistivity of the nematic liquid crystal containing ions. It can be written as $\rho = \lambda^2/(\varepsilon D)$, if the definition of λ is used [19]. It follows that equation (14) is equivalent to

$$\tau_r = \frac{\tau_{\mathrm{eq}}}{1 + (\delta\,\varepsilon)/(\lambda\,\varepsilon_S)} < \tau_{\mathrm{ion}}, \tag{15}$$

where τ_{eq} is given by equation (10) or equation (13), that, for the discussion reported above, are equivalent. The physical characteristics of the substrate entering in τ_r are the thickness, δ, and the dielectric constant, ε_S. From equation (15) it follows that the presence of the surface treatment always reduces the relaxation time of the nematic cell. This result is, a posteriori, evident. In fact, the presence of the surface layer introduces a capacitance in series to the one representing the nematic liquid crystal. It follows that the total capacitance of the cell is smaller than the one of the liquid crystal alone. Consequently, the relaxation time, defined as the product of the resistance of the nematic times the total capacitance, is reduced by the presence of the surface layer. The relaxation time given by equation (14) is compared with the experimental data reported by Sieberle and Schadt [8] for the nematic liquid crystal mixture 3825 oriented in a planar manner. The aligning coating is the polyamide CRC-6070-1 from Sumitomo Chemical. In this case $\varepsilon=5.26\varepsilon_0$ and $\varepsilon_S=3\varepsilon_0$, where ε_0 is the dielectric constant of the vacuum. The measurements were performed in the limit of small applied voltage. Figure 1 shows the dependence of τ_r vs. the thickness of the sample. The thickness of the surface layer is $\delta=0.15\,\mu\mathrm{m}$, in agreement with the reported value [8]. The best fit is obtained for $\rho=4.1\times10^8\,\Omega\,\mathrm{m}$, which is a reasonable value for the ionic conductivity of the liquid crystal. The dependence of τ_r vs. the δ is shown in figure 2. As reported by Sieberle and Schadt [8], $d=10\,\mu\mathrm{m}$, and the best fit is obtained for $\rho=1.1\times10^9\,\Omega\,\mathrm{m}$. Finally, in figure 3 we compare our predictions relevant to the dependence of τ_r with ρ. The agreement is not very good because we do not have precise values for ρ.

5. Influence of adsorption on the relaxation time

In the analysis presented above, by using the results obtained previously [19] on the equivalent circuit for a nematic liquid crystal cell obtained by solving the drift-diffusion equations for the ions dissolved in the sample, we have evaluated the relaxation time. In the analysis by Barbero and Alexe-Ionescu [19] the electrodes are supposedly completely blocking, and the basic equations of the problem are solved by assuming that the currents for the positive and negative ions, j_p and j_n, vanish on the surfaces limiting the sample. However, in some situations the limiting surfaces adsorb the ions. In this case, the boundary conditions by means of which it is necessary to solve the equations of continuity for the ions are of the type

$$j_\alpha = \pm \frac{\mathrm{d}\Sigma_\alpha^{\mathrm{ads}}}{\mathrm{d}t}, \tag{16}$$

on the limiting surfaces, where $\alpha=p$, n and $\Sigma_\alpha^{\mathrm{ads}}$ is the surface density of adsorbed ions. The kinetic equation

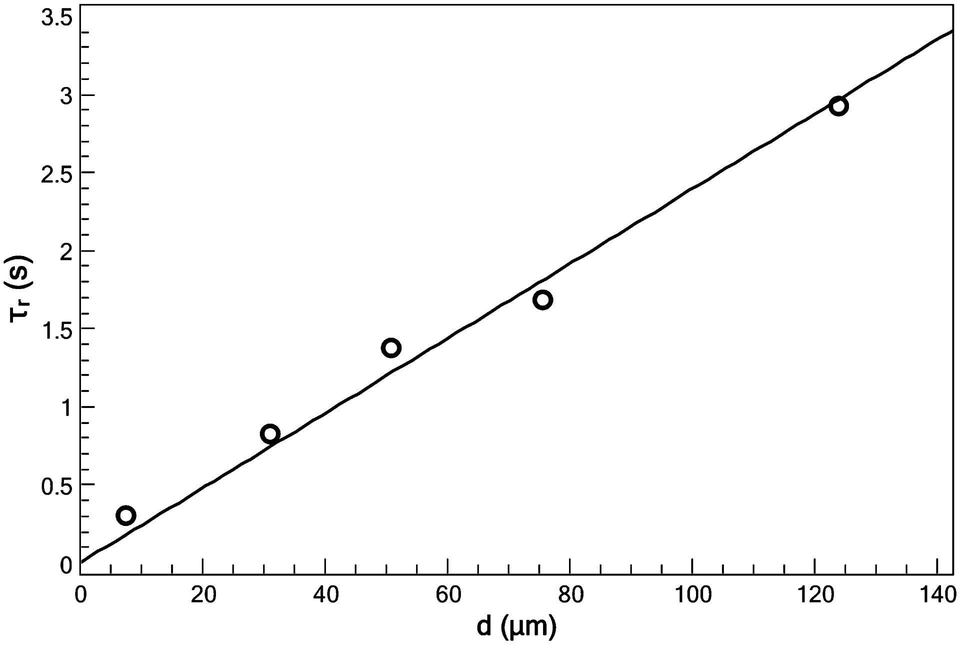

Figure 1. Relaxation time τ_r vs. the thickness of the sample d. The thickness of the surface layer is $\delta=0.15\,\mu\mathrm{m}$. The best fit is obtained for $\rho=4.1\times10^8\,\Omega\,\mathrm{m}$.

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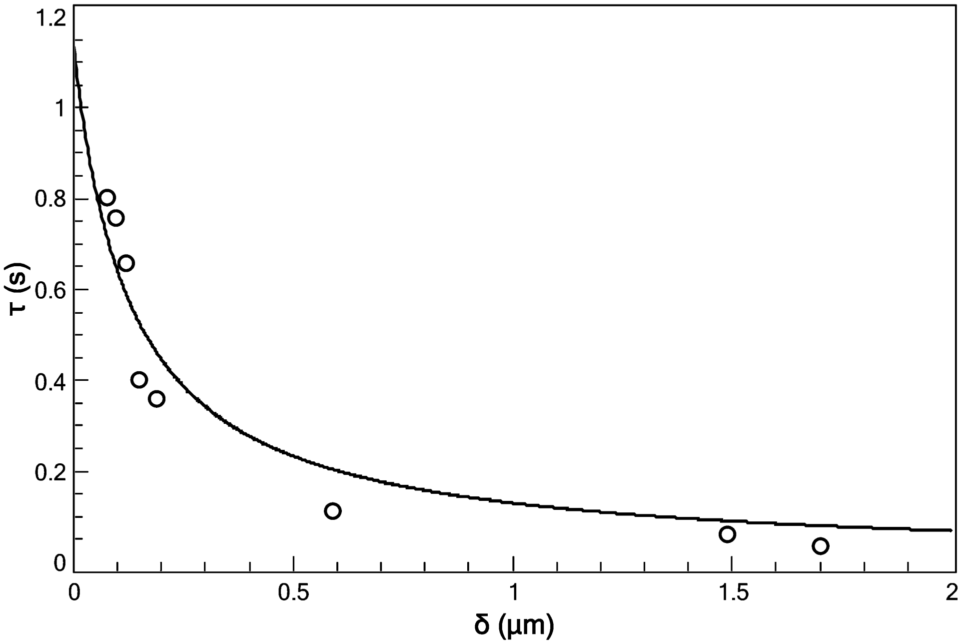

Figure 2. Relaxation time τ_r vs. the thickness of the surface layer δ. The thickness of the sample is $d=10\,\mu\mathrm{m}$. The best fit is obtained for $\rho=1.1\times10^9\,\Omega\,\mathrm{m}$.

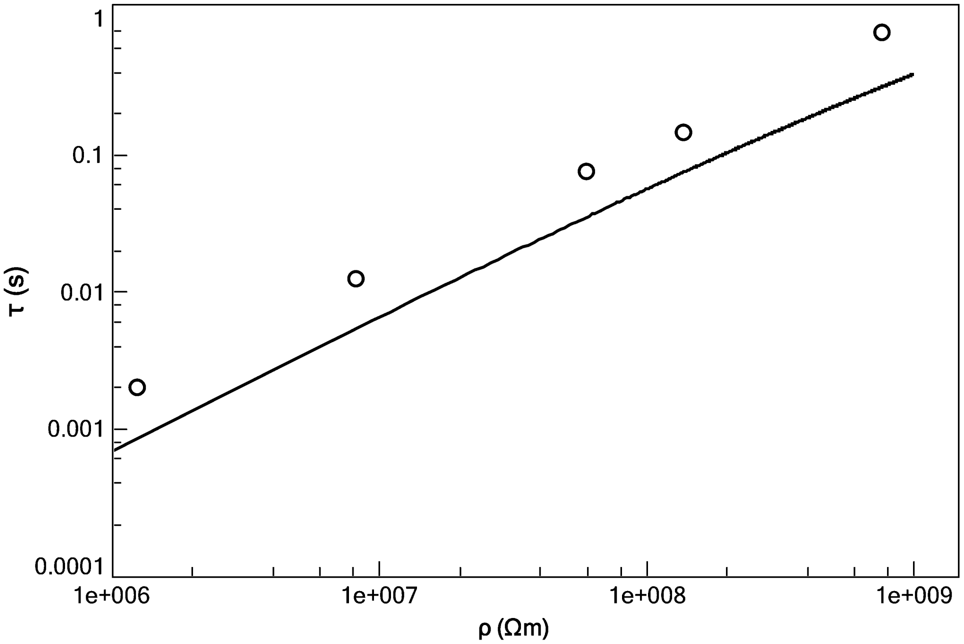

Figure 3. Relaxation time τ_r vs. the conductivity of the nematic material ρ. The thickness of the sample is $d=8\,\mu\mathrm{m}$ and the thickness of the surface layer $\delta=0.15\,\mu\mathrm{m}$. There are not free parameters.

for $\mathrm{d}\Sigma_\alpha^{\mathrm{ads}}/\mathrm{d}t$ is, usually, written in the Langmuir approximation, as

$$\frac{\mathrm{d}\Sigma_\alpha^{\mathrm{ads}}}{\mathrm{d}t}=k_{\mathrm{ads}}\rho_\alpha-\frac{1}{\tau_{\mathrm{ads}}}\Sigma_\alpha^{\mathrm{ads}}, \tag{17}$$

where k_{ads} and τ_{ads} are the adsorption and desorption coefficients, and ρ_α the bulk density of the ions α just in front of the considered electrode [3]. By means of an analysis similar to the one performed previously [19], it is possible to show that also in the case in which adsorption is present, in the low-frequency range, the equivalent circuit of the nematic cell is a series of a resistance and a condenser, as in the case considered above. The equivalent conductivity and dielectric constant in this case are no longer given by equation (12), but by [22]

$$\sigma_{\mathrm{eq}}^{\mathrm{ads}} = 2n_0\mu q\left\{1+\frac{D}{k_{\mathrm{ads}}d}\left(\frac{\eta}{1+\eta}\right)^2\right\}^{-1}, \qquad \varepsilon_{\mathrm{eq}}^{\mathrm{ads}} = \frac{1}{2}\varepsilon\frac{d}{\lambda}(1+\eta), \tag{18}$$

where $\eta = k_{\mathrm{ads}}\tau_{\mathrm{ads}}/\lambda$ is a dimesionless number characterizing the adsorption phenomenon. In this framework the dielectric relaxation time of the nematic, defined as before by $\tau_{\mathrm{eq}}^{\mathrm{ads}} = \varepsilon_{\mathrm{eq}}^{\mathrm{ads}} \big/ \sigma_{\mathrm{eq}}^{\mathrm{ads}}$, is

$$\tau_{\mathrm{eq}}^{\mathrm{ads}} = \tau_{\mathrm{eq}}(1+\eta)\left\{1+\frac{D}{k_{\mathrm{ads}}d}\left(\frac{\eta}{1+\eta}\right)^2\right\}, \tag{19}$$

where τ_{eq} is given by equation (13). When the substrate is considered, the effective relaxation time has to be evaluated as in the previous section. A similar calculation gives

$$\tau_r^{\mathrm{ads}} = \frac{\tau_{\mathrm{eq}}^{\mathrm{ads}}}{1+(1+\eta)(\delta\varepsilon)/(\lambda\varepsilon_S)}. \tag{20}$$

Also in this case, the presence of the surface treatment always reduces the relaxation time with respect to the one in which $\delta = 0$. We note that if the adsorption is weak, $\eta \to 0$ and the analysis presented in this section reduces to the one presented in the previous one. According to the measurements reported by Maximus *et al.* [3], $k_{\mathrm{ads}} \sim 10^{-7}\,\mathrm{m\,s^{-1}}$ and $\tau_{\mathrm{ads}} \sim 10^{-2}\,\mathrm{s}$. Assuming $\lambda \sim 0.1\,\mu\mathrm{m}$ [16], we get $\eta = 10^{-2}$ and $\tau_r^{\mathrm{ads}} \sim \tau_r$. It follows that in the measurements of relaxation time of the current in an electric circuit containing a nematic cell, the adsorption phenomenon is expected not to play a very important role. This is the probable reason why the agreement between the theoretical predictions of the model presented in the previous section, in which the adsorption phenomenon was neglected, and the experimental data [8] is rather good.

6. Conclusions

We have considered the relaxation of the electrical current in a circuit containing a nematic cell. We have shown that the analysis of the problem in terms of equivalent electrical circuits allows one to obtain a simple expression for the relaxation time that, however, cannot be used practically. The reason is that in the circuital analysis the dependence of the phenomenological parameters on the ionic density is not clear. By analyzing the ionic relaxation process in terms of drift-diffusion equations and Poisson's equation, we have determined the relaxation time for a single nematic material and for a nematic cell. Our analysis shows that the time relaxation of the nematic cell is reduced by the presence of the surface layer deposited on the electrodes to avoid charge injection and to orient the liquid crystal. Finally, we have considered the influence of the adsorption–desorption phenomenon on the relaxation time. According to our analysis, for the experimentally measured values of the adsorption and desorption coefficients, the influence on the relaxation time of this phenomenon is rather small.

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