

# TRANSIENT AND STEADY ELECTRIC CURRENTS IN A CELL WITH NEMATIC LIQUID CRYSTAL PENTYL-CYANOBIPHENYLE

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We have studied transient and steady electric currents in cells made of two glasses covered with indium tin oxide (ITO) and liquid crystal (LC) pentyl-cyano-biphenyl (5CB) in between. We demonstrated that space-charge effects are important here, contrary to what has been adopted before. Specifically, the nematic interlayer can be considered as an inhomogeneous medium with near-electrode double layers produced by some kind of a selective adsorption of ions supplied by the LC. The bulk of the LC contains charge carriers of both signs, which might be either extrinsic (impurity) or intrinsic ones. In the latter case, they can be imagined as heavy fragments of LC molecules. In the steady-current regime, the current-voltage characteristics are determined by the space charge injected or emitted into the nematic from the electrodes. For large enough biases  $U$ , the steady current saturates, since the space charge cloud is depleted. Transient phenomena observed by us testify that we are dealing with at least two kinds of charge carriers.

through and the complex dielectric constant of LCs in the low-frequency range allowed the mobilities and the diffusion coefficients of ions to be measured along and perpendicularly to the LC director, and the parameters of a double electric layer that arises at the electrode-LC interface to be evaluated.

The origin of ions that are responsible for currents in LCs still remains a matter of discussion. The authors of works [10, 11] suggested that inorganic ions play an important role in the formation of charge carriers in the nematic LC pentyl-cyano-biphenyl (5CB). The authors of work [12] suggested the positive polarity of the ions in 5CB and concluded that they mainly emerge due to a spontaneous dissociation of 5CB molecules. This conclusion was based on the fact that UV irradiation increased the ion concentration in 5CB, leaving the ion mobility the same.

## 1. Introduction

Despite the fact that LCs were discovered more than a century ago, the first systematic studies of the electric conductivity in a mesophase have not been started until the beginning of the 1970s, when a twist nematic mode was proposed for the implementation in LC displays (LCDs) [1–4]. The application of an ac electric field to an LC cell was found to excite a very weak transient current through the LC. Ionic impurities, which either are intrinsic to an LC matrix due to its chemical synthesis followed by a non-perfect purification or are a result of the LC contamination by the surrounding atmosphere, were suggested to be the origin of the transient current.

The transient current turned out to have a weak effect on the performance of the twist nematic mode. Therefore, little attention had been paid to studies of the electric current in nematic LCs, until active-matrix LCDs appeared in the market in the 1980s. Active-matrix displays required LC materials with exclusively low conductivity ( $< 10^{-12}$  S/m), and this circumstance gave impetus to a renewal of detailed studies of electric conductivity in LCs—in particular, nematic LCs (see, e.g., works [5–9]). The analysis of the transient current

Besides observing the transient electric current, the authors of work [13] reported that they revealed a weak steady current flowing through LC 5CB after the cell had been charged. The authors found that the steady current in a cell composed of two substrates covered with ITO electrodes and LC in between was proportional to the applied voltage only in the low-voltage regime, whereas nonlinear current-voltage characteristics (CVC) were observed, if the voltage across the cell exceeded a few volts. The magnitude and the very shape of the CVC turned out substantially dependent on the existence of an auxiliary aligning polyimide layer between the LC and the electrode. The authors of work [13] suggested the steady current across the cell to result from the charge-carrier recombination or a redox reaction at the electrode surface. They came to a conclusion that the current cannot be reduced to a space-charge-limited current only, although the CVC non-linearity favors the development of a space-charge cloud (see the description of corresponding phenomena in vacuum [14–16], gas-discharge or electron (ion) beam plasma [17–19], and insulating solids [20–22]).

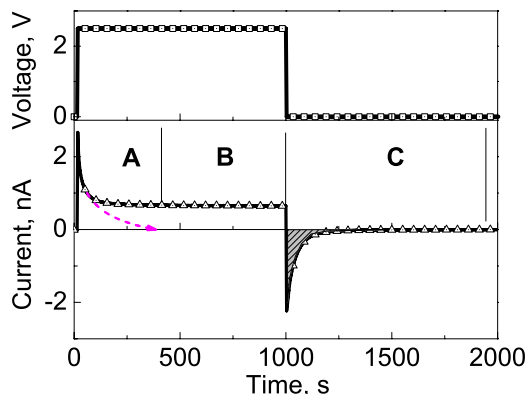


Fig. 1. Charging and discharging currents through a 20- $\mu\text{m}$  LC (5CB) cell between indium-tin-oxide (ITO) electrodes with the surface area  $S = 1 \text{ cm}^2$ . The applied bias voltage is  $U = 2.5 \text{ V}$

To our knowledge, the referred brief report [13] is the only one that informs about the observation of a steady electric current in nematic LCs. The flow of the steady current implies that electrode layers play an active role in the electric conductance of cells with nematic LCs at least for 5CB. Thus, while dealing with such problems, the whole industry of adsorption phenomena at relevant interfaces should be taken into account [23,24], including the influence—sometimes decisive—of image forces [25].

Charge transfer processes at the electrode–LC interface have become of special additional importance, because a number of surface-driven light-controlled effects have been found recently in nematic LCs [26–28]. These effects are caused by light-induced changes of the ion concentration in the vicinity of electrode–LC interface. Therefore, the response of an LC cell to light irradiation should be strongly affected by processes at the interface in the steady current mode.

In this paper, we describe our experiments aimed at studying the transient and steady electric currents across LC cells filled with 5CB. The corresponding analysis allowed intricate details concerning the formation of an electric double layer near the ITO surfaces and its characteristics to be revealed.

## 2. Experimental Part

We used LC cells composed of two glass substrates covered with ITO layers, and LC 5CB in between. The surface resistivity of ITO layers was  $R_s = 50 - 100 \Omega/\square$ . The area of ITO electrodes  $S = 1 \text{ cm}^2$  was less than the area of the corresponding sides of the LC slab. The LC thickness  $L = 20 \mu\text{m}$  was fixed by cylindrical polymer spacers deposited beyond the electrodes' areas. To avoid possible reorientations of LC by an external electric field,

we used the homeotropic orientation of LC on the ITO electrodes. The homeotropic alignment was achieved by the following treatment. First, the substrates were washed out with ethyl alcohol. After washing, they were rinsed in deionized water, the residuals of which were removed from the ITO surfaces by nitrogen blow-off. The whole treatment was performed at room temperature. Then, the LC cell was put into a grounded measuring unit connected with a pico-amperemeter; the latter can measure electric currents larger than  $10^{-15} \text{ A}$ . The applied dc voltage was controlled by a potential divider located in the measuring unit. The total resistance of the potential divider ( $20 \text{ k}\Omega$ ) was much lower than the resistance of the LC cell ( $2 \text{ M}\Omega$ ). The time-resolved analog signal from a pico-amperemeter was digitized by a data acquisition card.

## 3. Results and Discussion

The application of a dc voltage across the LC cell resulted in exciting a transient current  $I_t(t)$ , the relaxation time of which amounted to tens of seconds (Fig. 1, interval A). The current magnitude did not vanish in time, and reached a very small but reliably measured, finite steady value  $I_s$  of about 1 nA (Fig. 1, interval B). The steady current  $I_s$  did not change in time within several days. A short-circuit resulted in the emergence of a discharge current directed oppositely to the charge one (Fig. 1, interval C). It should be emphasized that a similar behavior was observed in nematic mixture E7, 5CB being one of its components. The value of the steady current was an order of magnitude less than that in 5CB. On the contrary, no steady current was observed for nematic mixture ZLI 4801, which did not contain cyano-biphenyl-based compounds (the discharge current through the cell filled with ZLI 4801 is depicted in Fig. 1 by a dotted curve). Similar extended transient currents were observed earlier for other insulating objects, namely, corona-charged polyethylene films [29].

The transient current is evidently caused by the polarization of the LC in a capacitor composed of ITO electrodes. The polarization originates from both bound and free charges in the cell. The bound charges are responsible, in particular, for the dielectric constants of pure 5CB ( $\epsilon_{\parallel} = 20, \epsilon_{\perp} = 18$ ). The charge  $q$ , which can be accumulated in our cell-capacitor due to the displacement of bound charges, is of the order of  $10^{-9} \text{ C}$  ( $\epsilon_{\perp} = 18, S = 1 \text{ cm}^2, L = 20 \mu\text{m}, U = 2.5 \text{ V}$ ). At the same time, the integration of the discharging curve depicted in Fig. 1 gives a much larger value,  $Q \approx 10^{-5} \text{ C}$ .

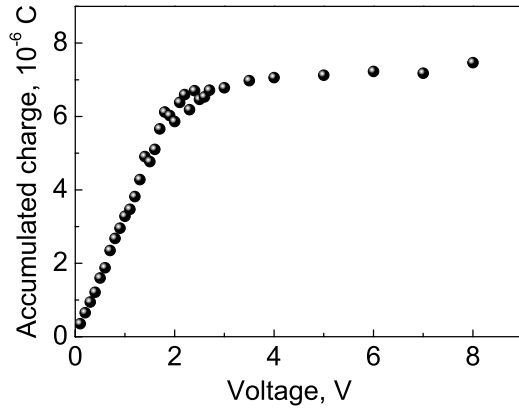


Fig. 2. Accumulated charge  $Q$  the versus bias voltage  $U$

Therefore, one can conclude that the redistribution of free charges in the double-layer region and in the cell bulk under the influence of an applied field gives the dominant contribution to the polarization of the LC cell.

The dependence of the charge  $Q$  accumulated in the cell on the applied voltage  $U$  is depicted in Fig. 2. This dependence was obtained by integrating the discharging transient current curve over the discharge time. It occurs that the accumulated charge  $Q$  increases approximately linearly up to  $U \approx 2$  V. Then a less steep, but also linear, growth of  $Q$  follows. The initial increase of  $Q$  is evidently caused by an enrichment of the near-electrode layers with accumulated charges. The formation of a charged double layer apparently terminates at  $U \approx 2$  V, and the subsequent slow increase of the accumulated charge is, probably, associated with the bound charge polarization or/and the exhaustion of the space-charge cloud.

Our hypothesis that the excess charge in the LC cell can be generated in either the charged double-layer region (both for the disconnected-circuit and steady-current regimes) or the space charge cloud stretched into the LC bulk is confirmed by the evaluation of the characteristic time of the excess charge formation. We measured the dependence of the accumulated charge  $Q$  on the time interval  $t_u$ , within which the voltage was applied (Fig. 3). The  $Q$ -values at different  $t_u$ 's were obtained by integrating the discharging transient current curve over the discharge time. The analysis of the experimental dependence  $Q(t_u)$  testifies that the single-exponential approximation

$$Q(t_u) = A_0 + A_1 \exp\left(-\frac{t_u}{\tau_1}\right), \quad (1)$$

where  $A_0$ ,  $A_1$ , and  $\tau_1$  are the fitting parameters, poorly describes the experimental dependence (dashed curve).

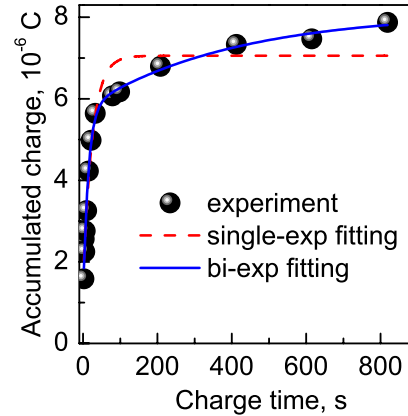


Fig. 3. Dependence of the accumulated charge  $Q$  on the charging time  $t_u$  and its single- and double-parameter fitting (see the text)

At the same time, the two-exponential approximation

$$Q(t_u) = A_0 + A_1 \exp\left(-\frac{t_u}{\tau_1}\right) + A_2 \exp\left(-\frac{t_u}{\tau_2}\right) \quad (2)$$

turned out much better (solid curve) and enabled us to evaluate the characteristic times of the formation of both the double layer ( $\tau_1 \approx 14$  s) and the space-charge cloud ( $\tau_2 \approx 397$  s) at  $U = 2.5$  V.

The dynamics of the cell discharge is governed by the diffusion of free carriers from the charged double layer into the bulk to restore the initial spatial distribution. The diffusion coefficient of carriers in the bulk,  $D$ , can be estimated by the characteristic decay time of the discharge transient current [31]. It turned out in our experiments that  $D \approx (2.5 \pm 0.3) \times 10^{-12}$  m<sup>2</sup>/s. The same value was obtained in the framework of the cell recharging method [32]. This value is by an order of magnitude lower than the self-diffusion coefficient of 5CB molecules,  $D = (6.5 \pm 0.5) \times 10^{-11}$  m<sup>2</sup>/s [33,34]. It corresponds to the hypothesis of the authors of work [12] on a “disassociation origin” of the steady current in 5CB. A decreased magnitude of the diffusion coefficient of charged 5CB ions in comparison to the self-diffusion coefficient of the neutral 5CB is naturally explained by the solvation of 5CB ions, resulting in an increase of the charge-carrier effective mass. It should be noted that the dielectric spectroscopy measurements made in works [9, 10] led to the values  $D = (1.9 \times 10^{-12} \div 1.2 \times 10^{-11})$  m<sup>2</sup>/s, so that the carriers responsible for the conductivity in 5CB were suggested to be positive and negative inorganic ionized impurities. The ion sizes were estimated to range from 0.25 to 1.5 times the Stokes radius, which gave the ground to the authors to presume those ions—naked, hydrated, or solvated by polar 5CB molecules – to be the majority charge

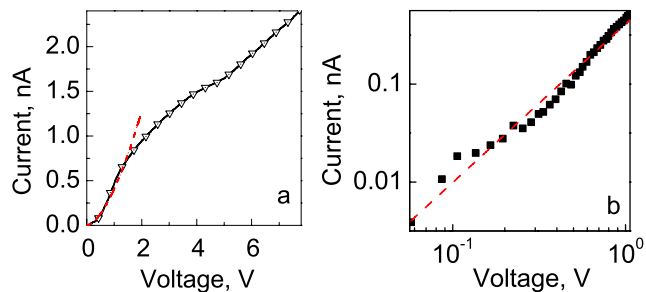


Fig. 4. (a) Experimental current-voltage characteristic of a steady current through the LC cell and its power-law fitting (dashed curve); (b) low-voltage section of the same dependences in log-log scaled-up coordinates

carriers in 5CB. In our opinion, the application of the Stokes approach to naked ions is at least doubtful, and it is reasonable to presume that very disassociated heavy 5CB molecules are responsible for the steady charge transfer in a cell filled with 5CB. Nevertheless, a contribution of impurity inorganic ions to the total conductivity can also not be excluded from consideration.

The concentration of 5CB ions in the cell can be evaluated by the formula

$$c_v = \frac{Q - q}{keSL}, \quad (3)$$

where  $q = CU$  is the bound charge accumulated in the cell at the applied voltage  $U$ ,  $C = \varepsilon_{\parallel}\varepsilon_0 S/L$  is the capacity of the cell with pure LC,  $e > 0$  is the elementary charge, and  $k$  is the ionization degree. For the actual geometrical parameters of the cell,  $U = 2.5$  V,  $\varepsilon_{\parallel} = 20$ ,  $Q = 6 \times 10^{-6}$  C, and provided  $k = 1$ , the average ionic concentration  $c_v \approx 2 \times 10^{16} \text{ m}^{-3}$ .

#### 4. Current-Voltage Characteristics

As was pointed out above, besides the traditional transient current, we observed the steady-state current  $I_s$  through our cell. Steady-state measurements were made at times after transient currents had decayed, namely, every CVC point was obtained in 43 seconds after the transition to a corresponding voltage value. The measured CVC for the steady current is presented in Fig. 4,a (solid curve). It demonstrates a strongly nonlinear behavior in the range of low and average applied voltages, and an almost linear dependence  $I_s(U)$  at  $U > 5$  V. We would like to repeat that a similar steady current was observed in nematic mixture E7, where 5CB is one of its components; but no steady

current takes place through nematic mixture ZLI 4801 free of cyano-biphenyl-based compounds. This means that free charges can appear only in the bulk of some LCs, although the double layer develops in any LC cell [23].

To gain some insight into the nonlinear CVC interval, one should pay attention to the fact that the very flow of the steady current across an insulator at low voltages implies the existence of a space charge with a finite density, which limits the magnitude of this current. Our estimations given above testify that rather a considerable extra charge ( $Q \approx 10^{-5}$  C) does exist in the LC cell bulk. This charge may hinder the current flowing through the cell and give rise to deviations of the CVC from the Ohmic behavior. Let us analyze the concave CVC section, bearing in mind a possible influence of the space charge, whatever its nature. We note that the conventional treatment of the charge motion driven by an external field through a vacuum gap or an insulating slab between metallic electrodes covers both the situation, where charge carriers are emitted into the slab through an energy barrier [14, 15], and the instance where the barrier is absent and carriers are injected into the space between electrodes [20, 21].

If ones neglects the thermal distribution of charge carriers over their energies (it does not change the dependence  $I(U)$  qualitatively [15, 35]), the CVC can be described analytically [14, 15, 17, 20]. To illustrate this, we restrict our scrutiny to two typical cases which might be realized in our LC cells. The first one has been originally introduced to describe a positive ion [17] or electron [14] inviscid flow in a vacuum gap between metallic electrodes. In this model, the CVC is determined by a system of three equations (see, e.g., work [15]),

$$\Delta V(\mathbf{r}) = -4\pi\rho(\mathbf{r}), \quad (4)$$

$$\mathbf{j}(\mathbf{r}) = \mathbf{v}(\mathbf{r})\rho(\mathbf{r}), \quad (5)$$

$$\frac{1}{2}m\mathbf{v}^2(\mathbf{r}) = eV(\mathbf{r}). \quad (6)$$

Formula (4) is the Poisson equation, where  $V(\mathbf{r})$  is the varying voltage in the gap at a point  $\mathbf{r}$  ( $V$  is reckoned from its cathode value, considered as zero), and  $\rho(\mathbf{r})$  is the charge density. Formula (5) is the relationship between the current density  $\mathbf{j}(\mathbf{r})$  and the charge velocity  $\mathbf{v}(\mathbf{r})$ , and  $m$  is the charge carrier mass. For simplicity, we assumed the thermal Maxwell distribution of charge carriers to be infinitely narrow. The third equation describes the electron (ion) acceleration in the

electrostatic field. As a consequence, the CVC follows the Child–Langmuir law at low voltages:

$$j = \frac{2}{9\pi L^2} \sqrt{\frac{e}{2m}} U^{3/2}. \quad (7)$$

Current (7) is limited by a space charge accumulated in the gap. At large  $U$ , the charge carriers are immediately driven away from the cathode, and the current tends to saturation. In particular, in the cold emission case,  $j(U)$  for large enough  $U$  obeys the Fowler–Nordheim law [15, 16]. At the same time, the form of the CVC for the space-charge limited current through the gas-discharge plasma [18, 19] is substantially influenced by collisions and collective excitations, the flow no longer being inviscid.

On the other hand, if charge carriers are emitted or injected into an insulator [20, 21], their motion is no longer accelerated and described by Eq. (6). Instead, one should take a material relationship between  $\mathbf{v}(\mathbf{r})$  and the electrostatic field  $\mathbf{F}(\mathbf{r}) = -\nabla V(\mathbf{r})$  into account:

$$\mathbf{v}(\mathbf{r}) = \mu(\mathbf{F}(\mathbf{r})), \quad (8)$$

where  $\mu$  is the charge carrier mobility (generally speaking, field-dependent). On the whole, Eq. (8) describes the charge carrier viscous drift in the external field  $\mathbf{F}(\mathbf{r})$  determined by collisions with impurities and imperfections. As a first approximation, we can neglect the diffusion current component, although it should be taken into account in the general case [20, 21].

Thus, in the model, where the insulator has no traps, polaron effects are absent, and  $\mu$  does not depend on  $\mathbf{F}(\mathbf{r})$ , the current density obeys the Mott–Gurney law at low  $U$  [20]:

$$j = \frac{9\varepsilon\mu}{32\pi L^3} U^2. \quad (9)$$

Here,  $\varepsilon$  is the dielectric constant of the isotropic insulator. One should expect that Ohm’s law  $j \sim U$ , appropriate to the charge flow in a resistive environment, is to be observed at large  $U$ ’s. If  $\mu = \mu(\mathbf{F})$ , the exponent power in Eq. (9) changes [21]. In particular, it might be equal to 3/2, as in Eq. (7), the nature of the conductance being different in both cases.

Looking at the experimental CVC (solid curve in Fig. 4,a), it becomes evident that, besides the Ohm behavior at high voltages, there also exist at least two other types of conductivity. One of them is actual at low voltages  $U < 1.5$  V, the other dominates in the intermediate  $U$ -range. It is natural that all the factors mentioned above alter the CVC, so that it can

be calculated only numerically. Nevertheless, one can consider Eqs. (7) and (9) as useful relations which provide some insight into the problem. Therefore, taking all the aforesaid into account, we attempted to describe the steady currents through our LC cells *at low voltages* by a power-law dependence  $I_s \sim U^\alpha$ , where  $\alpha$  is the fitting parameter. The results of calculations are shown in Fig. 4,b. The fitting curve is a linear dependence (solid line) in log-log coordinates with the slope  $\alpha \approx 1.64$ , whereas experimental points are denoted by black squares. Hence, the value of  $\alpha$  falls into the interval between 1.5 (Eq. (7)) and 2 (Eq. (9)). The observed deviation of  $\alpha$  from 2 means that the charge carrier mobility in the cell depends on the effective electrostatic field, i.e. on the applied voltage.

The fitting dependence is also depicted in Fig. 4,a (in the natural  $I - U$  co-ordinates) as a dashed curve. The success of our approximation means that it is quite plausible to consider the low- $U$  CVC region as that, where the current is limited by the space charge. At the same time, a conventional Ohm’s law is observed at large  $U > 4.5$  V, which is also in agreement with our arguments.

The remaining range of the CVC between 1.5 and 4.5 V reveals some tendency to saturation; so, it can be probably governed by some kinds of traps for moving ions [21]. Further experimental studies and theoretical analysis are needed to confirm this assumption.

We note that our experimental CVCs differ from those of Kovalchuk [36] obtained for cells with ZL1-4803-000 LC. He found an exponential dependence  $I_s(U) \propto \exp(\alpha\sqrt{U})$ ,  $\alpha$  being a constant, and interpreted it as either Richardson–Schottky attenuation [15] of the tunnel barrier located in the double layer near electrodes or Poole–Frenkel lowering [37] of the tunnel barrier for the carrier emission from charged traps in the bulk. It does not seem to be the case in our experiments.

## 5. Conclusions

To summarize, we have studied the transient and steady currents in nematic LC cells and demonstrated that the space-charge effects are important here, contrary to what was adopted before. Specifically, we conceive the nematic interlayer as an inhomogeneous medium with double layers near the electrodes produced by some kind of the selective adsorption of ions from the LC bulk. The bulk of the LC contains charge carriers of both signs, which might be either extrinsic (impurity) or intrinsic ones. In the latter case, they can be imagined as heavy fragments of LC molecules. In the steady-current regime,

CVCs are determined by the space charge injected or emitted into the nematic from the electrodes. For large enough biases  $U$ , the steady current is saturated, since the space charge cloud is depleted. The transient phenomena observed by us testify that we are dealing with at least two kinds of charge carriers.

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ПЕРЕХІДНИЙ ТА ПОСТІЙНИЙ  
СТРУМИ ЧЕРЕЗ КОМІРКУ, ЗАПОВНЕНУ  
НЕМАТИЧНИМ РІДКИМ КРИСТАЛОМ 5СВ

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Резюме

Вивчалися перехідний та стаціонарний струми через комірку, що складається з двох скелець, вкритих твердим розчином оксидів індію та олова, і заповнена рідким кристалом (РК) пентил-ціанобіфеніл (5СВ). Показано, що в цьому випадку ефекти просторового заряду є важливими, всупереч твердженням, що містяться в літературі. Зокрема, нематичний прошарок слід розглядати як неоднорідне середовище, в якому, внаслідок селективної адсорбції іонів з РК, утворюються приелектродні подвійні шари. В об'ємі РК знаходяться носії заряду обох знаків, що мають зовнішню (домішки) або внутрішню (великі фрагменти молекул РК) природу. В режимі стаціонарно-

го струму, вольт-амперні характеристики при малих напругах визначаються просторовим зарядом, утвореним внаслідок інжекції або емісії зарядів з електродів у немагніт. При достатньо великих напругах, вольт-амперні характеристики стають омич-

ними, оскільки накопичений просторовий заряд зменшується. Кінетичні характеристики перехідних процесів свідчать про те, що в данному випадку ми маємо справу щонайменше з двома типами носіїв струму.