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L. Tsonev; M. Petrov; G. Barbero

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# Electrical driving of the surface memorization in dimerized nematics with short range smectic order

L. TSONEV\*, M. PETROV

Institute of Solid State Physics, Bulgarian Academy of Sciences, 72 Tzarigradsko chaussee blvd., 1784 Sofia, Bulgaria

#### and G. BARBERO

Politecnico di Torino, Dipartimento di Fisica, c.so Duca degli Abruzzi 24, 18129 Torino, Italy

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Electrical control of the surface memorization of an oriented smectic C texture in the temperature range of dimerized nematics with short range smectic C order (4-*n*-alkyloxybenzoic acids) is presented. It is demonstrated that by suitable choice of the electric field parameters and surface conditions it is possible to separate the thermal and electrical components of the total erasure activation energy. The significant role of the double charge electric layer in the mechanism of the surface memorization is verified. By the electrical control of the memorization, we have confirmed the electrical part of the modified Rapini–Papoular anchoring energy in the effective surface energy.

#### 1. Introduction

Substrate interfaces which have an orienting action on liquid crystal (LC) molecules can be generated by a variety of means including chemical cleaning or etching, rubbing, SiO oblique evaporation, physical adsorption and deposition of organic or inorganic thin films. The surface-generated orienting forces may result from hydrogen bonding, physiochemical processes, van der Waals and/or dipolar interactions, or pure mechanical interactions induced by the liquid crystal anisotropic elasticity. So the balance of these forces, and therefore the resulting LC orientation, will be determined by the chemical composition of both the surface and the liquid crystal, as well as by the surface topology. Varying either of these conditions, the balance may be changed leading to change in the LC orientation [1].

Several theories have been proposed to explain the surface alignment of LCs: the Berreman model [2], the surface tension model [3, 4], van der Waals force model [5, 6], but these models cannot completely explain the experimental results for liquid crystal alignment. We have analysed the memorization (induced by the surface) of an oriented smectic C texture [7, 8] in the temperature range of the nematic phase and confirmed the idea that the investigation of this phenomenon is a useful method

for understanding the complex surface forces acting for different boundary conditions —various coatings and liquid crystal materials. The surface memory effect (SME), discovered by Friedel [9] has been studied in detail by Clark [10] who realized memorization of smectic A or C phases on hydrophobic polyimide surfaces in the nematic (and isotropic) temperature range. Some ideas for explanation of the SME and for understanding its mechanism were presented by Ouchi *et al.* [11] and Myrvold [12]. The mechanism of the SME, however, was still not clear. So far as the surface memorization is controlled by the temperature and by the surface conditions, the rotational diffusion model [11] proposed by Ouchi *et al.*, as well as the absorption model in [13] could be accepted as basic models.

Understanding of the surface memorization mechanism has become very important in recent years when it was found that the SME dominates the usual weak elastic anchoring in the 'flow alignment effect' [13]. In this case, the nematic anchoring on most of the isotropic or slightly anisotropic solid substrates is completely defined by the flow direction during the first contact of the nematic with the surface while filling the LC cell. Therefore, surface memorization could be studied from two aspects: (i) memorization of an oriented smectic texture in the nematic phase region, and (ii) 'flow alignment'. We discuss only the first aspect in the present paper.



<sup>\*</sup>Author for correspondence, e-mail: lyubo@issp.bas.bg

As we demonstrated in [7, 8], a quantitative study of surface memorization could be realized if one can control the two fundamental processes—recording and erasure. Until now such control was achieved by thermal excitation over the memorized texture (picture), inducing a counteracting torque proportional to the thermal

excitation over the memorized texture (picture), inducing a counteracting torque proportional to the thermal activation  $k_{\rm B}T$  in order to overcome the energy barrier of the irreversible deformation of the liquid crystal layer close to the surface. This thermal driving of the surface memorization structures enables us to apply the rotational diffusion model [11] to achieve a quantitative analysis of the SME. The fundamental parameters in this analysis are: the recording and the erasure temperatures  $(T_{\rm rec}, T_{\rm er})$ , the recording and erasure times  $(t_{\rm rec}, t_{\rm er})$ , the surface memorization strength  $\tau = t_{\rm er}/t_{\rm rec}$  and the erasure activation energy Q calculated from the dependence [7]  $t_{\rm er} = t_0 \exp(Q/k_{\rm B}T)$ , where  $k_{\rm B}$  is the Boltzmann constant and  $t_0$  is an extrapolation parameter. Knowing these parameters and the corresponding dependences, we have succeeded in presenting new information about the memorization mechanism.

We have demonstrated [7, 8] that physical adsorption is an important, but not the only mechanism of the SME. The total erasure activation energy Q is significantly bigger than the physical adsorption energy  $(Q_{ads} \approx 0.8 \text{ eV})$  due to the presence of some additional terms [8]:

$$Q = Q_{\rm ads} + Q_{\rm pol} + Q_{\rm mech} + \delta Q$$

where  $Q_{ads}$ ,  $Q_{pol}$ ,  $Q_{mech}$  and  $\delta Q$  will be clarified later in the text.

The purpose of the present work is to separate these terms by combining the standard thermal erasure process with an electrical excitation.

#### 2. Qualitative phenomenological analysis

One possibility for realizing the separation of  $Q_{ads}$ and  $Q_{pol}$  is the dependence found by us of the SME on the nature of the coatings—conducting or dielectric [8]. As we have found, some electrical phenomena (surface order electrical polarization and selective ion adsorption) are included in the surface memorization for dielectric and conducting surfaces, respectively. This inclusion is shown by the corresponding induced surface torques and the balance of these torques with the other surface and bulk forces during recording. In order to detect the electrical component of the SME, we excite the recording and the erasure process by external a.c. or d.c. electric fields. We took as driving parameters the frequency vand the amplitude U of the applied electric field and verified four possible fundamental electrical excitations depending on the experimental geometry and the liquid crystal materials-dielectric, electrohydrodyn amic, surface polar and flexoelectric—which can induce the corresponding instabilities. It is difficult to distinguish each one of these electric affects in a pure situation, but in our case, taking into account the geometry, the surface coatings and the dielectric anisotropy of the material used, by a microtextural polarization analysis, we found the dielectric and electrohydrodynamic excitations to be the most effective.

In order to illustrate the existence of different components in the erasure activation energy Q one can consider the total free energy density. The general presentation of the free energy includes different components [14]:

- (i) the bulk elastic energy  $f^{\text{elast}} = (K/2)(\partial^2 \theta / \partial r^2)$ , where K is the effective elastic constant and  $\theta = (\mathbf{n} \wedge \mathbf{n}_0)$  is the angle between the director **n** and its direction in the non-excited state  $\mathbf{n}_0$ ;  $\mathbf{n}_0$  is the 'easy' axis along which **n** would align on the surface in the absence of all other external fields;
- (ii) dielectric coupling with the electric field given by  $f^{\text{diel}} = -(\varepsilon_a/8\pi)(\mathbf{n} \mathbf{E})^2;$
- (iii) the surface energy  $f^{\text{surf}}$  which is generally assumed to be of the Rapini–Papoular form [15]:  $f^{\text{surf}} = -(W_{\text{s}}/2)(\mathbf{n}_{\text{s}} \mathbf{n}_{0})$ , where  $\mathbf{n}_{\text{s}}$  is the surface value of  $\mathbf{n}$  and  $W_{\text{s}} = K/L$ , L being the extrapolation length;
- (iv) the term  $f^{\text{surf.pol}} = -\mathbf{P}_{s}E\cos\theta_{s}, \theta_{s} = (\mathbf{n}_{s} \wedge \mathbf{n}_{0})$ , where  $\mathbf{P}_{s}$  represents that some surface polarization of a flexoelectric or ordoelectric nature is also possible [16].

Taking into account (a) the bulk and surface parts of the free energy density when the liquid crystal system is in an external (e.g. electric) field as indicated above; (b) the initial geometry ( $\mathbf{E} \perp \mathbf{n}_0$  or  $\mathbf{E} \parallel \mathbf{n}_0$ ; (c) the electric field parameters—voltage U and frequency v; (d) the material constants ( $\varepsilon_a$ —dielectric constant anisotropy and  $\sigma_a$ —electrical conductivity anisotropy), the possible bulk torques are [17]: dielectric bulk torque  $\Gamma_d \sim \varepsilon_a (\mathbf{E} \mathbf{n}_0)^2$ and the electrohydrodynamic torque  $\Gamma_{ehd} \sim \sigma_a E(\partial \theta / \partial r)$ . The corresponding surface torques participating in the balance equation are:  $\Gamma_s = -(K/\xi_E) \sin(\mathbf{n}_s \wedge \mathbf{n}_0)$ , where  $\xi_E^{-1} = (\varepsilon_a / 4\pi K)^{1/2} E$  and  $\xi_E$  is the electrical coherence length, and a stabilizing surface torque proportional to  $\sin(2\theta)$  [18].

It is also very important for the mechanism of the SME that a double electrical layer (see [8, 14]) exists at the interface conducting surface-liquid crystal due to the selective absorption of one type of ions (usually positively charged). Here we recall that in the dielectric component  $W_{el}$  of the effective anchoring energy  $W_{eff} = W_s + W_{el}$  there is included also the surface electric field  $\pm E_s$  which is confined in the double electrical layer

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induced by the selective ion adsorption. We can consider this electric field as internal with respect to the applied a.c. or d.c. electric fields.

If the molecules are polar, which is not so in our case, a surface polarization  $P_s$  can be induced and the corresponding surface polarization torque  $\Gamma_{\rm p} = -\mathbf{P}_{\rm s} E \cos \theta_{\rm s}$ . As mentioned,  $P_s$  could be a flexoelectric dipole or quadrupole type polarization. We shall present an experiment with dimer molecules where the quadrupolar aspect of the flexoelectricity could be active, but not the dipolar one. With the experiment which we present here, we cannot demonstrate the flexoelectric excitation either in the conducting or in the dielectric case. We found however that the SME does not exist in the case of a dielectric coating (glass, SiO, PVA). Recently we have found that using such surfaces [16] (SiO, obliquely evaporated) an ordoelectric surface polarization  $\mathbf{P}_{s}^{ord}$ could be detected. This polarization is due to the roughness of the solid SiO substrates since the scalar order parameter S decreases in the interface layer. As a result, the surface order parameter  $S_{\rm s}$  tends to zero or to a value very different from the bulk equilibrium value S(T). So the gradient of the nematic order parameter perpendicular to the surface over a distance  $\xi$  (correlation length)— $\nabla S/\xi$ —creates a horizontal (parallel to the substrates) 'ordoelectric' surface polarization  $P_s^{ord}$  which was indicated by a lateral electric field in  $\lceil 19 \rceil$ . Thus for the dielectric surface and  $\mathbf{E} \| \mathbf{n}_s$  two surface torques could be induced:  $\Gamma_x \sim E_x P_s^{ord} \cos \varphi$  and  $\Gamma_y \sim E_y P_s^{ord} \cos \theta$ where  $\varphi$  is the azimuthal angle of the **n**<sub>e</sub> director. The surface memorization on the dielectric surface is one of the two aspects of the SME [8] and the study by a lateral ( $\mathbf{E} \parallel \mathbf{n}_0$ ) electric field is very important in understanding the SME mechanism and as well as for the separation of the components of the erasure activation energy. However, we will develop this case in detail in future work. At this stage we will concentrate on the conducting surfaces, where according to our observations, surface polarization cannot be detected. In our erasure experiments however we observe dielectric and electrohydrodynamic regimes. For clarity we briefly recall the electro-optical characteristics of dielectric reorientation and the electrohydrodynamical instabilities in a nematic liquid crystal with short range smectic order and a small positive dielectric anistropy  $\varepsilon_a$  which is characteristic of the substance we use here. For details of these instabilities see [20].

The electrohydrodynamic instabilities (EHDI) in nematics with a different sign and magnitude of the dielectric anisotropy  $\varepsilon_a = \varepsilon_{\parallel} - \varepsilon_{\perp}$  and with an initial orientation of the director  $\mathbf{n}_0$  perpendicular to the applied electric field  $\mathbf{E}$  could be manifested as follows [20]: (i) at large negative values of  $\varepsilon_a$ , the nematic is stable for all electric fields and frequencies; (ii) EHDI is expressed in a 'conducting regime' (Williams domains) at low frequencies, and in a 'dielectric regime' at high frequencies. The two regimes are divided by a critical frequency  $v_c$ . Since we work with  $\varepsilon_a > 0$ , it is interesting that EHDI in this case can arise depending on the value of the anisotropy. Using the dependence  $E_{\text{th}}^2 = f(v^2)$  one can separate the EHDI and dielectric reorientation for a substance with  $0 < \varepsilon_a < \varepsilon_{a1}/3$ , where  $\varepsilon_{a1} = \varepsilon_{\parallel} \sigma_a / \sigma_{\parallel}$ . For example [20], for the substance used by us, 4-n-octyloxybenzoic acid with  $\varepsilon_a \approx 0.018$  and  $\sigma_a > 0$  at  $T - T_{NSc} \approx 2^\circ$ (i.e. in the nematic phase), the critical frequency  $v_c$  which clearly divides these two instabilities is  $v_c = 700 \text{ Hz}$ . Above this frequency only pure dielectric reorientation can be observed. The domain EHDI and dielectric instabilities are easily distinguishable by micropolarization analysis. The first one arises as cell-like domain lines, followed by dynamic scattering at v = const with increasing field U, where as the second one arises as a clear change of the colour, typical for the Fréedericksz transition.

Finally, let us mention the mechanical component of Q. It is difficult, however, to describe quantitatively this part  $Q_{mech}$  concerning the elastic adaptation of the director to the surface, when the surface topology is not well defined. That is why we give here only an estimation of  $Q_{mech}$  and propose a future experiment with a holographic grating as orienting surface, where the anchoring will be better defined. As is known from the Berreman model [2], the surface energy depends on the amplitude and the period of the grating. Changing these parameters, one can vary the surface energy value. In such a simplified experimental situation, a useful comparison between theory and experiment should be possible.

#### 3. Experiments and comments

We used cells with rubbed ITO coatings on the walls. The electric field was applied as  $\mathbf{E} \perp \mathbf{n}_0$  perpendicular to the substrates. The liquid crystal material was 4-*n*-octyl-oxybenzoic acid (OOBA) with the following temperatures (°C) of the phase transitions

$$Cr \xleftarrow{101} SmC \xleftarrow{108.1} N \xleftarrow{146} I$$

OOBA is a liquid crystal material involving hydrogenbonded dimer molecules. The electrical material constants of OOBA are as follows: in the SmC phase  $\varepsilon_a = -0.02$  and  $\sigma_a > 0$ ; in the nematic phase  $\varepsilon_a = 0.018$ and  $\sigma_a > 0$  [16]. The thickness *d* of the liquid crystal cells in our experiments varied between 3 and 200 µm.

The recording of surface memorized texture was realized by cooling the cell from the isotropic phase in the presence of an a.c. field down to the SmC phase. The recording temperature in the SmC phase was 105.5°C and the recording time  $t_{\rm rec} = 20$  min. The electric field was applied to stimulate formation of smectic C textures

characterized by large well oriented local single domains for the purpose of better observation and optical resolution. In table 1 the dependence of the SmC monodomains diameter on the a.c. electric field parameters is presented. The effective role of the electric field on the SmC local domain formation is, in general, readily seen: the domain size increases with increasing U at v = const, as well as with increasing v at U = const at low frequencies.

The dielectric or electrohydrodynamic textures were analysed by optical polarization microscopy. Varying  $T_{\rm er}$ ,  $v_{\rm er}$  and at fixed U we measured the erasure time  $t_{\rm er}$ . The results are presented in table 2. We must remember that for  $v = 5 \,\rm kHz$  the dielectric regime predominates, while for  $v < 700 \,\rm Hz$  the electrohydrodynamical regime predominates (see previous section).

The influence of an external a.c. field on the erasure time itself is obvious: the combined, simultaneous action of two destroying agents (the voltage used in our experiments for combined erasure is always over the instability threshold [20]) on the memorized texture shortens the erasure time compared with the purely thermal process. As is seen,  $t_{\rm er}$  tends to be considerably shorter with  $v_{\rm er}$  decreasing and for 145°C in the N phase,  $t_{\rm er} \approx 10$  s. The extreme values of the ratio  $t_{\rm er}^{\rm therm}/t_{\rm er}^{\rm el}$  at  $T_{\rm er} = 135^{\circ}$ C,

 $v_{\rm er} = 50$  Hz and  $d = 20 \,\mu{\rm m}$  is  $\approx 240/2 = 120$ . We concentrate our analysis on the erasure activation energy Q in the context of the general formula from the introduction.

Let us denote the energy Q for purely thermal erasure as  $Q^{\text{th}}$  and for combined erasure as  $Q^{\text{th}+\text{el}}$ . Our experiments (table 2) demonstrate that, in general,  $Q^{\text{th}+\text{el}} \neq Q^{\text{th}}$ and the deviation  $\Delta Q = Q^{\text{th}+\text{el}} - Q^{\text{th}}$  has different values and signs for different values of d and v. The most significant changes of  $Q^{\text{th}+\text{el}}$  with respect to  $Q^{\text{th}}$  are concentrated in the frequency interval 50 Hz-5 kHz; out of this interval  $\Delta Q$  is nearly constant and does not generate essential new information.

 $Q_{ads}$  and  $Q_{mech}$  are electrically independent;  $Q_{pol}$  (which includes for example the effect of the double electric layer [8, 14]) depends on the external electric fields of amplitude U and frequency v and could be modified by them in a combined erasure process. We can write:

$$Q^{\text{th}} = Q_{\text{ads}} + Q_{\text{mech}} + Q_{\text{pol}}(U = 0) + \delta Q$$
  
for purely thermal erasure

and

$$Q^{\text{th}+\text{el}} = Q_{\text{ads}} + Q_{\text{mesh}} + Q_{\text{pol}}(U, v) + \delta Q$$
for combined arcsure

for combined erasure

Table 1. Maximum effective diameter  $\Phi$  of SmC domains in textures which are recorded by cooling the nematic in the presence of a normal a.c. electric field of amplitude U and frequency v.  $d = 20 \,\mu\text{m}$ .

Parameter		Value										
U/V	40				80				160			
ν/kHz Φ/μm	0.2 30	1 30	2 10	5 5	0.2 100	1 300	2 800	5 50	0.2 150	1 300	2 500	5 100

Table 2. Erasure times  $t_{er}$  and erasure activation energies Q—thermal and under a.c. electric fields of different frequencies v—at fixed field amplitude U = 50 V and at different cell thickness values d in the case of OOBA confined between two ITO coated glass plates.  $t_{rec} = 20$  min,  $T_{rec} = 105^{\circ}$ C.

d/µm	$T_{ m er}/^{ m o}{ m C}$			a.c. electric erasure at frequency $v$						
		Thermal erasure		5kHz		500 Hz		50 Hz		
		$t_{\rm er}/{\rm min}$	$Q/\mathrm{eV}$	$t_{\rm er}/{\rm min}$	$Q/\mathrm{eV}$	$t_{\rm er}/{\rm min}$	$Q/\mathrm{eV}$	$t_{\rm er}/{\rm min}$	Q/eV	
6	145 140 135	5 10 30	2.6	3 6 14	3.6	2 5 10	2.3	1 2 5	2.3	
12	145 140 135	5 8 19	2.2	3 7.5 20	3.2	3 6 11	2	1.5 3 6	2.0	
20	145 140 135	20 60 240	4	5 30 90	4.2	1 3 6	3	0.2 1 2	1.8	

where  $\delta Q$  describes other possible mechanisms of memorization anchoring (e.g. steric interactions) and

$$Q_{pol}(U, v) = Q_{pol}(U = 0) \quad [1 + f(U, v)];$$
  
the function f is always  $f \ge -1$ .

When the external field weakens the electrical connections described by  $Q_{pol}(U=0)$ , then  $Q_{pol}(U, v)$  becomes lower than  $Q_{pol}(U=0)$ . In the extreme 'reduction' case  $Q_{pol}(U, v) = 0$ , i.e. f = -1, therefore

$$Q^{\text{th}+\text{el}} - Q^{\text{th}} = -Q_{\text{pol}}(U=0) = -|\Delta Q|_{\text{max}}.$$

When the external field strengthens the electric connections described by  $Q_{pol}(U=0)$ , then  $Q_{pol}(U, v)$  becomes larger than  $Q_{pol}(U=0)$ . Even in the extreme 'positive' case, however, the increase of  $Q_{pol}$  cannot be higher than  $Q_{pol}(U=0)$  itself, i.e.  $f \le 1$  and

$$Q^{\mathrm{th}+\mathrm{el}} - Q^{\mathrm{th}} = Q_{\mathrm{pol}}(U=0) = |\Delta Q|_{\mathrm{max}}$$

Our main assumption is that the electrically determined component  $Q_{pol}$  of the erasure energy  $Q^{th}$  does not exceed the maximum experimentally measured deviation  $|\Delta Q|_{max}$ , i.e.

$$Q_{\rm pol} \leq |\Delta Q|_{\rm max}$$

Practically, we have (see the figure and table 2): (i) for cells of thickness  $d \neq 20 \,\mu\text{m}$ :  $|\Delta Q|_{\text{max}} = 1 \,\text{eV}$ ,  $Q^{\text{th}} = 2.2 \div 2.6 \,\text{eV}$ ; for cells of thickness  $d = 20 \,\mu\text{m}$ :  $|\Delta Q|_{\text{max}} = 2.2 \,\text{eV}$ ,  $Q^{\text{th}} = 4 \,\text{eV}$ . Therefore we propose the following phenomenological estimation: the electrical contribution  $Q_{\text{pol}}$  in  $Q^{\text{th}}$  does not correspond to more than half of the erasure activation energy  $Q^{\text{th}} \equiv Q$ , i.e.

$$Q_{\rm pol} \leq Q/2.$$

Up to now we do not develop a full end exact model of the SME; we suppose that such an estimation is accept-



able, at least to give an idea about the order of magnitude of  $Q_{pol}$  by separating it in this way from the other components of Q.

Let us summarize the results for the purely thermal erasure process:

$$Q = Q_{ads} + Q_{mech} + Q_{pol} + \delta Q$$
$$Q_{pol} \approx Q/2$$
$$Q_{ads} \approx 0.8 \text{ eV (typically, see e.g. [7])}.$$

Therefore we find another estimation:

$$Q_{\rm mech} + \delta Q \approx Q/2 - Q_{\rm ads}$$

In our case: (i) for cells of thickness  $d \neq 20 \,\mu\text{m}$ ,  $Q_{\text{mech}} + \delta Q \approx 0.5 \,\text{eV}$ ; (ii) for cells of thickness  $d = 20 \,\mu\text{m}$ ,  $Q_{\text{mech}} + \delta Q \approx 1.2 \,\text{eV}$ . Therefore we can reach another important conclusion:

$$Q_{\rm mech} < Q_{\rm pol}.$$

For some parameters of the electric field and  $\mathbf{E} \perp \mathbf{E}_s$ , the dependence from the figure disappeared, i.e. the external electric field depressed the thickness variation of Q (table 2), meaning a variation of the ratio  $q_s/q_b$ where  $q_s$  and  $q_b$  are the surface and bulk free charges, respectively, thus eliminating the electric part  $W_{\rm el}$  of the surface effective energy  $W_{\rm eff} = W_s + W_{\rm el}$  in the case of conducting substrates, as a result of electrical double layer depression. In such a way we confirmed our assumption in [8] that in the surface memorization mechanism, the role of selective ion adsorption and the creation by it of a double electric charge layer is significant.

We have also observed that the mean single local monodomain diameter decreases when the cell thickness decreases. This fact obviously correlates with the decrease of the erasure activation energy and of the SME strength below  $20 \,\mu\text{m}$ . The cell thickness of approximately  $20 \,\mu\text{m}$  is critical in this sense. So the dependence in the figure presented here for the memorization strength seems to be very general for the surface phenomena at the interface between dimerized nematics and solid substrates. A similar dependence was found for the scalar order parameter and anchoring energy thickness dependence [21] for dimerized nematics, as well as for the dependence of the selective ion density on the cell thickness for the classical nematics presented by Barbero *et al.* [22].

#### 4. Conclusions

An electrical control of the surface memorization of the oriented smectic C texture in the temperature range of dimerized nematics with short range smectic C order (4-*n*-alkyloxybenzoic acids) is presented. It is demonstrated that by suitable choice of the electric field parameters and surface conditions it is possible to separate



the thermal and the electrical components of the erasure activation energy. The significant role of the electrical double charge layer in the mechanism of surface memorization is verified. By the electrical control of the memorization we have confirmed selective ion adsorption and the electrical part of the modified Rapini–Papoular anchoring energy in the effective surface energy.

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