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Mechanism of nematic molecular alignment based on friction charges and surface topology by rubbing

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A new and simple electric charge interaction mechanism has been proposed to explain the behaviour of the tilt angle of liquid crystals by rubbing. The mechanism of the molecular alignment is elucidated based on the effect of a static electric charge on a substrate surface treated by rubbing. An electric field which is dependent on the topology of a substrate surface allows a planar orientation of molecules with positive dielectric anisotropy. With increasing rubbing strength, the tilt angle varies slowly. The director of molecules with negative dielectric anisotropy is uniform and has a tilt angle determined by the molecular permanent dipole direction. The air–liquid crystal interface case is also considered. This model enables us to give a unified picture of the molecular alignment mechanism.

1. Introduction

In the practical applications of liquid crystals (LC) in display devices, one of the most important problems encountered is how to obtain uniform alignment of the LC molecule. The unidirectional rubbing [1], SiO oblique evaporation [2], or coating of surfactants [3] performed on the cell are techniques commonly used at present. In these alignment techniques, the rubbing method is usually used in the display devices. In spite of the practical convenience of this alignment techniques, the mechanism of LC molecule alignment at the substrate surface and in a bulk is still not well understood. The interfacial properties of a substrate and a LC are also not known well enough to allow an unambiguous description of the alignment mechanism.

In this paper the main subject is to undertand the molecular alignment mechanism in a nematic LC cell with polyimide layers induced by a rubbing. The mechanism of the molecular alignment is explained based on the effect of static electric charge on a substrate surface treated by a rubbing. An electric field which is induced by static electric charges dependent on a topology of substrate surface allows a planar orientation of molecules with positive dielectric anisotropy. With increasing rubbing strength, the tilt angle varies slowly [4]. On the other hand, the director of molecules with negative dielectric anisotropy is uniform and has a tilt angle determined by the molecular permanent dipole direction. The temperature dependence of the tilt angle as well as the influence of the thickness of the polyimide layer will be explained by the coupling of the LC molecular dipole and the friction charge. These results are in agreement with experiments. In this theory the air–LC interface, i.e. the free surface case is also considered. The results again agree with the theoretical prediction.

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2. Electric field by friction charges

Although electricity produced by friction was known in ancient times, a quantitative description is still lacking for this phenomenon. Therefore our discussion starts with a simple introduction about charging polymers. Given the present state of knowledge concerning polymers, one cannot determine exactly the frictional charge induced by the rubbing on the polymeric substrate. Therefore a simple assumption that the polymeric film possesses a uniform surface charge is taken as a first step toward understanding this phenomenon. This simple assumption is appropriate for both the model mechanisms mentioned above.

To treat the effect of the surface topography we limit consideration to a nematic sample with a grooved substrate surface [5] as shown in figure 1. We also assume that there is a surface density of friction charge denoted by σ_0. In figure 1 the LC lies in the region $y>0$, and the polymer layer is in $-\xi \leqslant y < 0$ where ξ is the thickness of the polymeric substrate. By assuming $Aq<1$, the electric field induced by the surface charge can be represented as the composition of two fields. The first field is caused by a uniform surface charge σ_0 at the $y=0$ plane. The second field is caused by a surface dipole density at the same plane.

At first we consider the first field whose corresponding potential $\varphi(y)$ should satisfy the Laplace equation, $d^2\varphi(y)/dy^2=0$, in both regions of $y>0$ and $y<0$. The boundary condition is shown by $D(y=0^+)-D(y=0^-)=4\pi\sigma_0$, where $D(y=0^+)=-\varepsilon^{\mathrm{L}}[d\varphi(y)/dy]|_{y=0^+}$ and $D(y=0^-)=-\varepsilon^{\mathrm{P}}[d\varphi(y)/dy]|_{y=0^-}$ are the electric displacement corresponding to $\varphi(y)$ in both sides of the plane $y=0$, and ε^{L} and ε^{P} are the dielectric constants of the LC and the polymer, respectively. From the boundary condition that the total potential of the two field are zero at $y=+\infty$ and $y=\xi$, we solve $d^2\varphi(y)/dy^2=0$ and obtain

$$\varphi^{\mathrm{L}}(y>0)=\text{const.}=4\sigma_0 A(\varepsilon^{\mathrm{P}}-\varepsilon^{\mathrm{L}})/\varepsilon^{\mathrm{L}}\varepsilon^{\mathrm{P}}, \tag{1}$$

and

$$\varphi^{\mathrm{P}}(y<0)=\frac{4\pi\sigma_0}{\varepsilon^{\mathrm{P}}}(y+\xi). \tag{2}$$

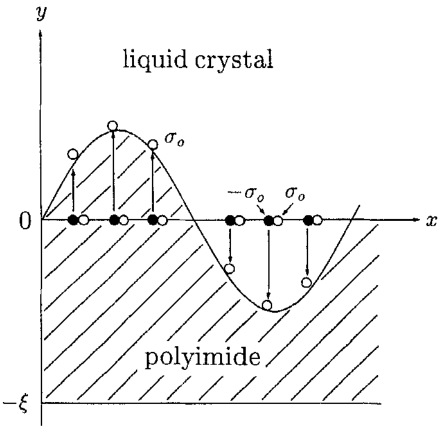

Figure 1. Schematic representation of the contact between a liquid crystal and a grooved substrate surface described by $y=A\sin(qx)$ [5] in which A is the amplitude of the groove and q ($2\pi/q$ is wavelength of the groove) is the wavenumber. The rubbing direction is normal to the sheet. Circles on a groove show the friction true charges by rubbing and pair circles at $y=0$ plane show virtual charges.

The second field, i.e. the dipole field, needs a more complex derivation. The potential of the field is denoted to be

$$\psi=\begin{cases}\psi^{\mathrm{L}}(x,y) & (y\geqslant 0),\\ \psi^{\mathrm{P}}(x,y) & (-\xi\leqslant y\leqslant 0).\end{cases} \tag{3}$$

The interfacial boundary condition is given by

$$\psi^{\mathrm{L}}(x,0)-\psi^{\mathrm{P}}(x,0)=\begin{cases}\dfrac{4\pi P}{\varepsilon^{\mathrm{P}}} & (0<\sin qx<1)\\[2ex] \dfrac{4\pi P}{\varepsilon^{\mathrm{L}}} & (-1<\sin qx<0)\end{cases}$$

$$=\frac{4\pi\sigma_0 A}{\varepsilon^{\mathrm{P}}\varepsilon^{\mathrm{L}}}\left[\frac{\varepsilon^{\mathrm{L}}-\varepsilon^{\mathrm{P}}}{\pi}+\tfrac{1}{2}(\varepsilon^{\mathrm{P}}+\varepsilon^{\mathrm{L}})\sin qx+\frac{2(\varepsilon^{\mathrm{P}}-\varepsilon^{\mathrm{L}})}{\pi}\sum_{n=1}^{\infty}\frac{\cos 2nqx}{4n^2-1}\right]. \tag{4}$$

where $P=\sigma_0 A\sin qx$. Over a dipolar surface the electric potential must jump $4\pi\sigma_0 A/\varepsilon$, where ε is the local dielectric constant of the dipolar position. This condition is true in a meaning of the planar capacitance. Another interface condition which comes from the Gauss theorem specific for a dipolar surface is the continuity of the normal component of **D** which is given by

$$\varepsilon^{\mathrm{L}}\frac{\partial}{\partial y}\psi^{\mathrm{L}}(x.\,0)=\varepsilon^{\mathrm{P}}\frac{\partial}{\partial y}\psi^{\mathrm{P}}(x,0). \tag{5}$$

The potential of the second field is satisfied with the Laplace equation

$$\frac{\partial^2}{\partial x^2}\psi+\frac{\partial^2}{\partial y^2}=0. \tag{6}$$

Considering the same boundary conditions at $y=+\infty$ and $y=-\xi$ as the first field the general solution is given by

$$\psi=\begin{cases}\psi^{\mathrm{L}}(x,y)=\displaystyle\sum_{n=1}^{\infty}\exp(-nqy)(a_n\cos nqx+b_n\sin nqx)+a_0,\\[2ex] \psi^{\mathrm{P}}(x,y)=\displaystyle\sum_{n=1}^{\infty}\sinh[nq(y+\xi)](c_n\cos nqx+d_n\sin nqx).\end{cases} \tag{7}$$

The value shown in equation (1) just balances a_0 to keep the relation of $\varphi^{\mathrm{L}}(y=+\infty)+\psi^{\mathrm{L}}(y=+\infty)=0$. By combining two fields the total potential $(V^{\mathrm{L}}=\varphi^{\mathrm{L}}+\psi^{\mathrm{L}})$ in LC bulk is given for the region of $y\geqslant 0$

$$V^{\mathrm{L}}=\frac{2\pi\sigma_0 A(\varepsilon^{\mathrm{P}}+\varepsilon^{\mathrm{L}})}{\varepsilon^{\mathrm{L}}(\varepsilon^{\mathrm{P}}+\varepsilon^{\mathrm{L}}\tanh q\xi)}\exp(-qy)\sin \mathrm{qx}$$

$$+\sum_{n=1}^{\infty}\left[\frac{8\sigma_0 A(\varepsilon^{\mathrm{P}}-\varepsilon^{\mathrm{L}})}{\varepsilon^{\mathrm{L}}(\varepsilon^{\mathrm{P}}+\varepsilon^{\mathrm{L}}\tanh 2nq\xi)}\right]\frac{\exp(-2nqy)\cos 2nqx}{4n^2-1}. \tag{8}$$

This equation shows that the orientation field is proportional to the A and σ_0. This means that the field responds to the rubbing strength.

3. Orientation effect

Now we suppose that the rubbed polyimide affects the molecular orientation due to the charging field given by equation (7). To verify this prediction we now analyse the orientation effect for the field.

In order to calculate the field we take the operation of a gradient on V^{L} and obtain the electric field in LC bulk

$$\mathbf{E}^{\mathrm{L}} \equiv [E_x^{\mathrm{L}}, E_y^{\mathrm{L}}] = -\left[\frac{\partial}{\partial x}, \frac{\partial}{\partial y}\right] V^{\mathrm{L}}.$$

It is useful to calculate the mean field of $\mathbf{E}^{\mathrm{L}}$. In figure 1 we divide each complete periodic region into two separated parts. The integration of $\mathbf{E}^{\mathrm{L}}$ for the two parts give the resultant mean fields for each part

$$\bar{\mathbf{E}}^{\mathrm{L}} = \frac{4\sigma_0 A(\varepsilon^{\mathrm{P}} + \varepsilon^{\mathrm{L}}) q \exp(-qy)}{\varepsilon^{\mathrm{L}}(\varepsilon^{\mathrm{P}} + \varepsilon^{\mathrm{L}} \tanh q\xi)} \times \begin{cases} [0, 1] & (\sin qx > 0) \\ [0, -1] & (\sin qx < 0) \end{cases}. \tag{9}$$

This implies that the higher order terms and the component in x direction disappear. By assuming the LC orientation with tilt angle $\theta(y)$ along to the rubbing direction the director is defined by $\mathbf{n} = (n_x, n_y, n_z) = (0, \sin\theta(y), \cos\theta(y))$. The LC total free energy [6] is

$$F = \int g\,dv = \int \left[\frac{1}{2}k\left(\frac{d\theta}{dy}\right)^2 - \frac{1}{8\pi}\Delta\varepsilon^{\mathrm{L}} M^2 \exp(-2qy)\sin^2\theta - \frac{1}{8\pi}\varepsilon_{\perp}^{\mathrm{L}} M^2 \exp(-2qy)\right] dv, \tag{10}$$

where $M = [4\sigma_0 A(\varepsilon^{\mathrm{P}} + \varepsilon^{\mathrm{L}})q]/[\varepsilon^{\mathrm{L}}(\varepsilon^{\mathrm{P}} + \varepsilon^{\mathrm{L}} \tanh q\xi)]$, and dv is the volume element.

3.1. *For the case of* $\Delta\varepsilon^{\mathrm{L}} < 0$

The minimization of F gives a unique solution, $\theta = \theta_0$, where θ_0 is the pretilt angle dependent on the total anchoring at LC–substrate interaction. The energy includes not only the van der Waals [7] and steric interactions [8] but also the present rubbing charge effect. The latter effect may be simply thought as the interaction between the field and the present molecular dipole at the surface

$$g^{\mathrm{S}} = -\boldsymbol{\mu}_0 \bar{\mathbf{E}}^{\mathrm{L}}, \tag{11}$$

where $\boldsymbol{\mu}_0$ is the dipole moment on the surface. Hence, the pretilt angle θ_0 must be affected by the rubbing. In the negative anisotropy case, the molecular dipole is almost perpendicular to the molecular long axis, i.e. $\mathbf{n}$. Obviously, from equations (9) and (11), the rubbing field tends to reduce to θ_0, i.e. forming a planar alignment. The field takes alternative signs with the space as shown in equation (9) which also ensures that this is no polar alignment at the surface as required for a nematic phase.

3.2. *For the case of* $\Delta\varepsilon^{\mathrm{L}} > 0$

The corresponding Euler–Lagrange equation to minimize F is obtained

$$k\frac{d^2\theta}{dy^2} + \frac{\Delta\varepsilon^{\mathrm{L}}}{8\pi} M^2 \exp(-2qy)\sin^2\theta = 0. \tag{12}$$

This is a nonlinear differential equation and its integration cannot be solved analytically. In order to see the feature of the orientation as easily as possible we solve the following appropriate equation

$$k\left(\frac{d\theta}{dy}\right)^2 - \frac{\Delta\varepsilon^{\mathrm{L}} M^2}{4\pi}\exp(-2qy)\sin^2\theta = 0. \tag{13}$$

Physically the condition of equation (13) keeps the balance of the elastic and electric energies [9]. Therefore the solution of equation (13) shows the same tendency as that of equation (12), and may be seen a reasonable approximation of that of equation (12).

The term of $\sqrt{(k/\Delta\varepsilon^{\mathrm{L}}M^2)}$ has the dimension of length. Now the new parameter as the characteristic length of the rubbing charge effect is introduced by

$$\xi_M=\frac{2}{M}\sqrt{\left(\frac{\pi k}{\Delta\varepsilon^{\mathrm{L}}}\right)}. \tag{14}$$

Using this definition equation (13) changes to

$$\xi_M^2\left(\frac{d\theta}{dy}\right)^2=\exp(-2qy)\sin^2\theta. \tag{15}$$

There are two solutions in equation (15) which is satisfied by

$$\frac{d\theta}{dy}=\pm\exp(-qy)\frac{\sin\theta}{\xi_M}. \tag{16}$$

By integrating equation (12) and assuming the strong anchoring condition of $(d\theta/dy)|_{y=0}=0$ we have

$$\frac{d\theta}{dy}=-\frac{1}{4\xi_M^2}\int_0^y\exp(-2qy)\sin^2\theta\,dy<0. \tag{17}$$

This leads to one choice for equation (16). By selecting the branch with negative sign, its integration gives.

$$\frac{1+\cos\theta(y)}{1-\cos\theta(y)}=\left(\frac{1+\cos\theta_0}{1-\cos\theta_0}\right)\exp\left[\frac{2}{\xi_M q}(1-\exp(-qy))\right], \tag{18}$$

where θ_0 is the pretilt angle at the surface $y=0$. $\theta(y)$ is always decreasing with y. In other words the competition between the elastic and rubbing charge forces again results in the tendency of planar orientation as a negative case. However, one should note that there is a little difference: the real tilt angle or the so called effective pretilt angle in the case of $\Delta\varepsilon>0$ should be defined by $\theta_0'=\theta(y\to\infty)$, as

$$\frac{1+\cos\theta_0'}{1-\cos\theta_0'}=\left(\frac{1+\cos\theta_0}{1-\cos\theta_0}\right)\exp\left[\frac{2}{\xi_M q}\right]. \tag{19}$$

Equation (19) shows clearly that if $\xi_M q$ decreases then θ_0' also decreases. From the definitions of M and ξ_M, one finds that $\xi_M q$ is almost independent of q, but depends on $A\sigma_0$ and material parameters only. This result follows the experimental result [10] that in the rubbing technique the groove shape is relatively unimportant, but the strong rubbing (i.e. larger $A\sigma_0$) will reduce the tilt angle.

3.3. *For the case of free surface*

There is another branch of the solutions to equation (13) (i.e. the positive one in equation (16)). This is just opposite to the behaviour of equations (17) and (18), and cannot describe the rubbing effect at a solid surface. However, it is an equilibrium state and might have its use for other cases. A free surface of liquid crystal is a very suitable case in which the 'grooves' come from natural fluctuation and the charges are caused by absorption in air. Then one can expect that in the LC bulk $\theta(y)$ will increase with y based on

$$\frac{1+\cos\theta(y)}{1-\cos\theta(y)}=\left(\frac{1+\cos\theta_0}{1-\cos\theta_0}\right)\exp\left[\frac{-2}{\xi_M q}(1-\exp(-qy))\right]. \tag{20}$$

Then the alignment opposite to the planar one happens. In fact homeotropic alignment at the free surface is firmly observed for 5CB, 7CB and 8CB [11–12]. We believe that more precise measurement may show their tilt angle θ'_0 not to be exactly 90° which is given by the relationship at the limit of $y \rightarrow \infty$, $(1+\cos\theta'_0)/(1-\cos\theta'_0) = [(1+\cos\theta_0)/(1-\cos\theta_0)]\exp(-2/\xi_M q)$.

4. Discussion

The above discussions for both the cases of $\Delta\varepsilon < 0$ and $\Delta\varepsilon > 0$ show that homogeneous alignment can be always achieved by the rubbing regardless of the original molecular orientation being parallel or tilted on the untreated surface. However, one should note from equation (19) that the original tilt θ_0 has its resistance to rubbing effect. Experiments [13] show that it is very difficult to get the homogeneous alignment when the LC molecules originally align perpendicular to the nonrubbed surface. Figures 2 (*a*–*c*) summarize the three types of LC orientation discussed for $\Delta\varepsilon < 0$ ((*a*)) and $\Delta\varepsilon > 0$ ((*b*) and (*c*)) which includes the rubbing treatment and the free surface, respectively. The obvious difference between $\Delta\varepsilon < 0$ and $\Delta\varepsilon > 0$ in the rubbing treatment is the director behaviour. For the case of $\Delta\varepsilon < 0$ the director is uniform while a little distortion appears in the case of $\Delta\varepsilon > 0$.

We showed that the well-known homogeneous alignment of the LC by rubbing can be explained by the friction charge field. For further support of this model it is necessary

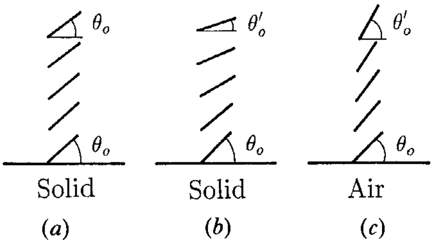

Figure 2. Three types of liquid crystal orientation: (*a*) for the case of $\Delta\varepsilon < 0$, (*b*) for the case of $\Delta\varepsilon > 0$ in which the liquid crystal is in contact with a rubbing substrate surface, and (*c*) for the case of $\Delta\varepsilon > 0$ in which the liquid crystal is in contact with air.

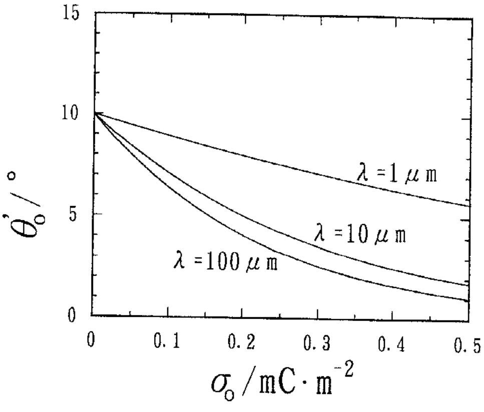

Figure 3. Friction charge density, σ_0, dependencies of surface tilt angle, θ'_0, as a parameter of assumed wavelength with $A = 10$ nm and $\theta_0 = 10°$.

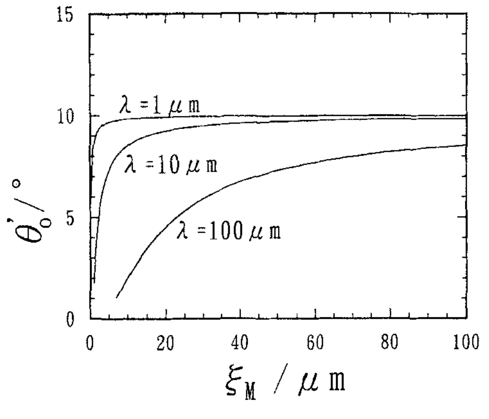

Figure 4. Relations between ξ_M and θ_0' with $\theta_0 = 10°$.

to provide more direct evidence. As is proposed in the theory the tilt angle θ_0 (for $\Delta\varepsilon < 0$) or θ_0' (for $\Delta\varepsilon > 0$) can be reduced by the rubbing field $\bar{E}^L$ shown in equation (9). In equation (9) the polyimide thickness is included in a part of denominator with the functional form of $\tanh(q\xi)$. It is then easy to find that the strength of $\bar{E}^L$ decreases with increasing the thickness ξ, but saturates at $\xi \to \infty$ due to the characteristics of $\tanh(q\xi)$ function. Accordingly, the tilt angle θ_0 or θ_0' will increase with ξ, but has the same saturation property. The result on the influence of substrate polymer thickness is quite significant for fundamental and practical interest. For the application of liquid crystal displays performance, this is a useful technology to control the tilt angle.

The σ_0 dependencies of θ_0' at the constant $\theta_0 = 10°$ for the case of $\Delta\varepsilon > 0$ are calculated using equation (19) to see the influence of the friction charges on the tilt angle as shown in figure 3. Figure 4 shows the calculated results of the relation between ξ_M and θ_0' for various wavelength of the groove using equations (14) and (19). It is clear that the characteristic length ξ_M which is introduced in this study and is determined by the rubbing strength affects the tilt angle for the case of $\Delta\varepsilon > 0$. In these calculations the constant value of $\sigma_0 = 0{\cdot}25\,\mathrm{mC\,m^2}$ is used. This value is reported [14] as the contact charge density of a polyimide with a nylon which is usually used as a rubbing brush. The friction charge density may not be larger than the contact charge density. A full account of the analysis, with the temperature and the polymer film thickness dependence of the tilt angle, will be published in a forthcoming paper [9].

5. Conclusions

A new and simple charge interaction mechanism has been proposed to explain the behaviour of the tilt angle between the nematic LC director and a substrate treated by rubbing and other methods. This model has enabled us to give the molecular alignment mechanism a unified treatment to explain why rubbing can always give the planar orientation and also the temperature and substrate thickness dependences of the tilt angle [9]. There would appear to be three types of tilt alignment: one is the uniform tilt for a LC with negative dielectric anisotropy; second for director distortion from the pretilt angle to the effective one, and third at the LC free surface which assumes the reverse behaviour to the second type. The contributions to the tilt angle may be divided into two parts: at the surface, the pretilt angle is determined by a balance between van der Waals forces and the rubbing or adsorption charge field, and the bulk orientation near the surface is the result of competition between the same charge field and the curvature elastic torque of the LC. Because knowledge of the van der Waals alignment force and its effects on the pretilt angle is still lacking, this paper is limited to giving a detailed analysis for the strong anchoring assumption. For the bulk case, however, the knowledge of a tilt angle will be sufficient for study.

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